

Composition, interactions and resulting inhalation risk of micro- and nano-plastics in urban air

Corresponding Author: Professor Hartmut Herrmann

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Decision Letter:

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Dear Professor Herrmann,

Your manuscript titled "Micro- and nano-plastics (MNPs) in urban air: polymer composition, interactions and inhalation risk" has now been seen by 3 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

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

Additionally, the following editorial thresholds need to be addressed:

*Provide compelling novel insight into the quantification strategies while assessing the potential health risks through inhalation of airborne micro- and nanoplastics (MNPs) in urban air

*clearly mention the limitations/uncertainties associated with the methodological analyses including discussion on 'one site sampling' and 'risk assessment while exposed to urban air'.

Please submit your point-by-point responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

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

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

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

been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

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

Best regards,

Suryasarathi Bose, PhD
Editorial Board Member
Communications Earth & Environment
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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This study investigated the microplastic mass concentrations within inhalable air at an urban location using a Pyrolysis GCMS technique, finding that tire wear particles were the largest contributor to PM10 mass of all the polymers targeted. With large discrepancies in the reported values of airborne microplastic concentrations, this study is highly relevant to the field, and it is a notable advancement in our understanding urban air pollution and the potential health impacts. The study has very strong analytical merit, using the most up to date methodologies and considerations. I recommend it for publication after addressing minor comments (below).

Comments:

Methods: Overall, the pyrolysates used in quantification are judiciously selected. However, I have a question related to the choice for TTT: 2,4-Dimethyl-4-vinylcyclohexene. It has been reported by Li et al. (2023) [<https://doi.org/10.1016/j.jaap.2022.105816>] that HCl produced from PVC pyrolysis can react with vinyl functional groups of other pyrolysates, leading to final values that may be underreported. Did the authors notice any secondary reactions related to 2,4-Dimethyl-4-vinylcyclohexene or are they able to make a comment on the potential for secondary reactions?

Were the authors able to find any correlations of tire wear particles with the amount of traffic in the city or observe a trend in the amount of tire wear in weekend vs weekday samples?

Line 201 – it would be helpful to the reader if the term “by mass” was added when referring to the percentages.

Line 256 (and Supp Table 6) – please define OC, EC, TC, POC, SOC, POM when they are first introduced in the text.

Line 423 – please provide information on what solvents were used for the dissolution of each polymer.

Line 425 – a one point calibration was used for C-PP and C-PE, a method which typically cannot be reported as quantitative. I note that the cited reference Goßmann et al. explains the semi-quantitative nature of using the 1-point calibration methodology. If dissolution of the PE and PP was not possible, did the authors try to dilute in sand or another matrix when preparing calibration standards? More discussion on the limitations of a one-point calibration method is needed.

Supp Fig 1 – It would be helpful if the authors could list the equations for each calibration curve.

Line 519 – Was a laboratory fume hood used or a laminar flow hood for the drying procedure? A typical fume hood will draw in air from the surrounding laboratory, so there would still be risk for contamination. Please comment. It would also be beneficial to the reader if the authors could provide the average amount (and standard deviation) of plastics found in the blanks, as it is noted that blank subtraction was performed (line 532).

Line 522 – It was noted that nitrile gloves were used, however, it has been shown that the use of nitrile gloves can lead false positives in Pyrolysis GCMS experiments [Witzig et al. (2020) [<https://doi.org/10.1021/acs.est.0c03742>]]. Did the authors observe any contamination peaks in the resulting pyrograms associated with use of nitrile gloves?

Reviewer #2 (Remarks to the Author):

General comments:

This paper presents the concentration of airborne micro- and nano-plastic particles over a two week period in an urban environment in Germany. The analysis was done for PM10-2.5 (coarse MP; CMP) and PM2.5 (fine MP; FMP), along with the total MNPs. In addition, total, elemental and organic carbon were assessed from the same filters. The analytical methods are solid and represent the current state of the art for MNP (especially in the smaller size ranges). The results suggest that tire wear particles are the dominant form of airborne MNP at this urban, roadside site, followed by other common polymers: PVC, PE, PP, etc. The authors present correlation matrices to evaluate the pairwise coincidence of polymers and carbon, showing

that many of the polymers are correlated with carbon. This isn't surprising, given that the MNPs are likely measured as part of the carbon (albeit as a small proportion of the total carbon), and that when particulate loads are high all types of particle abundance will increase. The more novel aspect of the study comes from the application of inhalation risk analysis, where the authors demonstrate an enhanced risk associated with exposure to urban air.

The greatest short-coming in the paper is that only one site was sampled, so any variability in the concentration of the particles is likely to come from day-to-day patterns in weather or traffic, rather than any clear variation in particle source. There was no presentation of weather variability over the study period, however, so it is unclear what caused the small variation in concentrations and relative abundance of polymers and sizes. Thus, the correlations presented don't provide as much insight as desired, since there is so little variability in the data. Perhaps a multivariate analysis (NMDS or similar) would be more helpful in illustrating patterns rather than multiple bivariate correlations. This would be a much stronger contribution if the study had been carried out at multiple sites with different potential sources, even if only at different levels of traffic or urban activities within the same city.

The manuscript is generally well written, but there are a number of places where the writing was confusing, or perhaps errors in editing. The Results/Discussion section is quite long, with a lot of detail on correlations or potential health impacts of polymers that could be removed or shortened to better streamline the results. The results and the figures are high quality and generally illustrate the patterns well. I suggest considering a different color scheme for figures 1b, 3, S3, etc because the shades of blue and green are so close that they are indistinguishable and the red/green combination may be challenging for some readers.

Specific comments

Abstract

Line 21 – fine and coarse MP should be defined here

Line 24 – it would help to indicate briefly which polymers were most common

Line 25 – it's not clear that the data support "complex atmospheric interactions" given the lack of variability in the dataset or any investigation of mechanisms

Line 26 – the polymer-specific hazard index should be defined here, or the statement simplified to avoid confusion by the reader

Introduction

Line 35 – a more typical definition of microplastic is <5 mm, so limiting to <1 mm should be explained or justified using a reference

Line 39 – this sentence mixes the matrix where MP were found with the region/location and should either be limited to region, or indicate the matrix for all regions

Line 45 – consider rewording: "In spite of increase prevalence in the environment" and explaining what is meant by "specific single-source contribution". The latter is confusing, as MP sources are highly variable

Line 49 - ... into the deep lung where they are suspected to cause

Line 51 – remove one mention of "airborne"

Line 54 – remove "There are"

Line 59 – airborne MNPs are under-represented

Line 68 – estimates with a polymer-specific

Line 88+ - Is this only true for PVC, or for other MP polymers?

Line 111 – what were the weather conditions during the study and did they vary? Does this variability have any relation to the observed patterns?

Line 126 – Were any statistical analyses done to evaluate the patterns, aside from the correlation analysis?

Line 134 – smaller than other polymers? Is it preferential, or just a characteristic of the polymer that smaller MPs are released from the original product?

Line 148+ - this section reads a bit like a list of results from other studies and could synthesize these comparisons a bit more

Line 148 – In this section, several sentences are worded as "City reported high values". I find this wording awkward, as the cities themselves aren't reporting the values. Consider "higher levels of PE were reported for Kyoto, Japan...", etc

Line 156 – this statement that methodological variability limits intercomparison is very important, and a note about which methods were used for the studies cited may be warranted if there were differences in size fractions analyzed, instrumentation, extraction, etc

Line 174 – it may be more clear to indicate that there were two size classes and the total, since the total is the sum of the FMP and CMP

Line 181 – remove "the" before following

Line 186 – this sentence is hard to follow. I'm not sure what the "indicates" is referring to

Line 189 – this relationship could be evaluated statistically by doing a pairwise comparison between polymer abundance between the two size fractions and also by presenting a multivariate analysis of the two size classes to evaluate potential drivers of the overall composition of MP in each

Figure 2 – would it be easier to see the polymers with lower abundance if the CTT and TTT were removed, or if the other polymers were blown up to better illustrate the composition of the less abundant materials?

Line 201 – how was the total mass load measured? This isn't clear from the methods, unless I've missed something. I see the methods for the polymer analysis and the EC/OC analysis, but not total mass.

Line 201+ - this section could also be more synthetic rather than listing other studies

Line 230+ - likewise here, where this section could better synthesize the patterns, however, the lack of variability in the dataset may make further interpretation difficult, as it doesn't appear that there is much change in weather over the time period and the data are from a single source. For example, why does the widespread use of PE, PET and PS in commercial

products explain why they correlate with one another? The Wright study cited here used weather data to evaluate potential source areas of different materials, and the Chen study in Shanghai correlated MP patterns with traffic and weather data.

Line 245 – this conclusion about co-occurrence of road markings and tire particles is logical, but without evaluating sources that may vary in potential loading of these materials it is harder to draw this conclusion

Line 255 – is this statement suggesting that the carbon sum parameters are indicative of adsorbed C on the surface of the FMP? If the MP are such a small proportion of the total PM load, and also a small proportion of atmospheric particulate C, it seems that it might be challenging to detect adsorption of materials in bulk on fine relative to coarse MP particles?

Line 259 – This statement about CMP dominance needs clarification

Line 263 – is this statement suggesting that PMMA, PVC, and PE are part of the POC and SOC pools? Or is there another meaning here?

Line 285 – I'm not certain that the data support this statement entirely. The correlations are based on a single site, and any variability may be due to simple shifts in traffic or wind that kick up particles, so that the abundance of all particles in the atmosphere will increase concomitantly. Is there evidence that the MPs are aggregated directly with or adsorbed to the carbonaceous particles? Or just that they co-occur?

Line 296+ - this section had a number of sentences that were hard to follow, and perhaps were editing errors, especially line 304, 307, 310, 311

Line 304 – this statement seems to be mixing routes of exposure between airborne and food-based sources

Line 313 – it isn't clear which findings are referred to here – the present study or the Mumbai study?

Line 340+ - the hazard categories (line 342), associated chemicals (lines 347+) and potential human health impacts (lines 353, 355, 356, etc) could be put into a table that summarizes the potential toxicity associated with prior studies, and this section streamlined to focus more on the data in hand and which materials may pose the greatest risk

Line 383 – space after comma

Line 405 – the sentences here appear incomplete

Line 410 – were filtered

Line 430 – check capitalization

Line 442+ - I believe that the PM_{2.5} and the PM₁₀ were measured separately on the two different instruments deployed in the field, but this could be made more clear

Line 506 – by background levels, do you mean the levels of PM without microplastic?

Reviewer #3 (Remarks to the Author):

This work provides a comprehensive analysis of airborne micro- and nanoplastics (MNPs) in urban air, highlighting tire wear particles (TWPs) as the dominant source and quantifying their potential health risks through inhalation exposure. The use of Py-GC-MS for polymer-specific characterization is robust, and the integration of carbonaceous aerosol interactions adds valuable insights. However, limitations include methodological ambiguities (e.g., C-PVC* cluster interpretation), insufficient discussion of comparative studies, and overextrapolation of health risks without polymer-specific toxicological validation.

Specific comments:

1. Clarify how C-PVC* cluster differentiation from black carbon (BC) was handled, given its mixed signals (Page 4-5). Statistical methods for correlation analysis (e.g., p-values) are missing in Supplementary Table 6.
2. Discrepancies in MNP concentrations (e.g., Shanghai: 5.6 µg/m³ vs. Leipzig: 0.6 µg/m³) lack discussion on methodological differences (sampling, analysis) or emission source variations (Page 6).
3. Extrapolating PM_{2.5} risk models to MNPs requires justification, as polymer-specific toxicity (e.g., C-PVC* PHI) may not align with generic PM toxicity (Page 18-19).
4. Justify why Torgauer Straße (high traffic) was chosen as the sole sampling site. Urban background vs. roadside variability should be addressed (Page 23).
5. One-point calibration for C-PE/C-PP (low concentrations) is inadequate. Provide detection limits and error margins (Page 22).
6. Excluding C-PA6/MDI-PUR due to "undetectable signals" (Page 5) may bias results. Discuss potential underestimation of total MNP mass.
7. Daily intake calculation assumes fixed outdoor exposure time (4h). Validate with local activity patterns or sensitivity analysis (Page 24).
8. SDG linkages (Page 20) are generic. Specify actionable policies (e.g., tire wear regulations, MNP monitoring standards).
9. Abbreviations (e.g., FMPs/CMPs/TWPs) are defined multiple times (Abstract, Section 2.1.1). Consolidate definitions in the Introduction.
10. Contrast Py-GC-MS with other techniques (e.g., µFTIR, Raman) in Introduction/Discussion to highlight methodological advantages.

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Version 1:

Decision Letter:

<*** REMEMBER TO ATTACH REVISIONS CHECKLIST (WORD)***>

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Dear Professor Herrmann,

Your manuscript titled "Micro- and nano-plastics (MNPs) in urban air: polymer composition, interactions and inhalation risk" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

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

Best regards,

Suryasarathi Bose, PhD
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Nandita Basu, PhD
Associate Editor, Communications Sustainability
Consulting Editor, Communications Earth & Environment
Nature Portfolio
<https://www.nature.com/commssustain/>

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have addressed all comments clearly, and I believe the manuscript is now in a publishable form.

-Joel Rindelaub

Reviewer #2 (Remarks to the Author):

This is a very nice revised manuscript and I appreciate the thorough response to comments and questions. Wonderful work!

Reviewer #3 (Remarks to the Author):

All the concerns have been well addressed, it can be accepted now.

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Comments

This work provides a comprehensive analysis of airborne micro- and nanoplastics (MNPs) in urban air, highlighting tire wear particles (TWPs) as the dominant source and quantifying their potential health risks through inhalation exposure. The use of Py-GC-MS for polymer-specific characterization is robust, and the integration of carbonaceous aerosol interactions adds valuable insights. However, limitations include methodological ambiguities (e.g., C-PVC* cluster interpretation), insufficient discussion of comparative studies, and overextrapolation of health risks without polymer-specific toxicological validation. **Recommendation: MINOR REVISION.**

Specific comments:

1. Clarify how C-PVC* cluster differentiation from black carbon (BC) was handled, given its mixed signals (Page 4-5). Statistical methods for correlation analysis (e.g., p-values) are missing in Supplementary Table 6.
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regulations, MNP monitoring standards).

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10. Contrast Py-GC-MS with other techniques (e.g., μ FTIR, Raman) in Introduction/Discussion to highlight methodological advantages.

All the concerns have been well addressed, it can be accepted now.

Response to Reviewers for the manuscript of "Micro- and nano-plastics (MNPs) in urban air: polymer composition, interactions and inhalation risk" by A. Kaushik et.al.

We thank reviewers for their comments which we think are most helpful to further improve our submission. Please see below point-by-point answers, addressing the reviewer's recommendations. We are listing the original comments **in red**, our replies in black and any changed or amended text **in blue**. Line numbers refer to the continuous line numbering of the revised manuscript.

Reviewer: 1

Recommendation: Publication after addressing minor comments.

Comments:

This study investigated the microplastic mass concentrations within inhalable air at an urban location using a Pyrolysis GCMS technique, finding that tire wear particles were the largest contributor to PM₁₀ mass of all the polymers targeted. With large discrepancies in the reported values of airborne microplastic concentrations, this study is highly relevant to the field, and it is a notable advancement in our understanding urban air pollution and the potential health impacts. The study has very strong analytical merit, using the most up to date methodologies and considerations.

We sincerely thank the reviewer for these positive remarks and for recognizing the analytical robustness and relevance of our study. We appreciate the acknowledgement of our work's contribution to understanding the composition and potential health implications of airborne micro- and nano-plastics (MNPs) in urban environments. We have strived to employ state-of-the-art Pyrolysis-GC-MS methodology and comprehensive data interpretation, and we are pleased that the reviewer finds these aspects to be a notable advancement in the field.

R1C1: Overall, the pyrolysates used in quantification are judiciously selected. However, I have a question related to the choice for TTT: 2,4-Dimethyl-4-vinylcyclohexene. It has been reported by Li et al. (2023) [<https://doi.org/10.1016/j.jaap.2022.105816>] that HCl produced from PVC pyrolysis can react with vinyl functional groups of other pyrolysates, leading to final values that may be underreported. Did the authors notice any secondary reactions related to 2,4-Dimethyl-4-vinylcyclohexene or are they able to make a comment on the potential for secondary reactions?

A-R1C1: We appreciate the reviewer's insightful observation and for drawing attention to the findings of Li et al. (2023). The current study quantifies CTT and TTT using already published quantifiers in Goßmann et al., 2023 (<https://doi.org/10.1016/j.scitotenv.2021.145667>). In addition to that, in our measurements, we did not observe distinct additional peaks or altered chromatographic patterns in the TTT marker region that would suggest notable secondary

reactions between HCl from PVC pyrolysis and 2,4-Dimethyl-4-vinylcyclohexene. We also considered other secondary reactions which were reported by Coralli et al., 2022 (<https://doi.org/10.1016/j.iaap.2021.105377>). Secondary reactions during the pyrolysis process are becoming much less relevant in highly diluted and also natural, complex samples. They are known almost exclusively with respect to artificial experimental setups with selected compounds present in comparable high concentrations. The much higher dilution factor and the presence of multiple reactants for these specific pyrolysis products (e.g., HCl) suggest that these reactions occur to a negligible extent, if at all. No corresponding indications have emerged in the course of the analyses. However, they cannot be completely ruled out. These particular aspects we have highlighted in the revised version of the manuscript to make it clearer.

So, we have amended the manuscript text as follows:

page 24-25, line 491-507: MNPs analysis was performed as outlined in published literature^{19,38,39,66,104} by using Py-GC-MS. The instrument set-up consisted of a micro furnace pyrolyzer (EGA/Py-3030D; FrontierLabs, Japan) with an auto-shot sampler (AS1020E; FrontierLabs, Japan), which was coupled with a GC-MS (6890N-5973MSD; Agilent Technologies). All the quantifiers have been chosen by taking into consideration the secondary reactions that may happen during the pyrolysis process^{105,106}, but these reactions are largely irrelevant in highly diluted, complex environmental samples. Such reactions are reported mainly in artificial experimental setups where selected compounds are present at relatively high concentrations. In natural samples, the much higher dilution and the presence of multiple reactants for potential pyrolysis products (e.g., HCl) suggest that these reactions occur only to a negligible extent, if at all. Our analyses showed no evidence of such effects, although they cannot be completely excluded. Moreover, the measurement was performed in Single Ion Monitoring (SIM) mode, which enhances the sensitivity of the target compound by focusing on characteristic ions of target polymers (Supplementary Table 3). This also reduces background noise from complex airborne matrices and allows reliable quantification of polymers at trace levels, providing lower detection limits and improved accuracy compared with full scan analysis.

Reference added to the revised manuscript:

Goßmann, I., Halbach, M. & Scholz-Böttcher, B. M. Car and truck tire wear particles in complex environmental samples – A quantitative comparison with "traditional" microplastic polymer mass loads. *Science of The Total Environment* 773, 145667 (2021).

Li, B. et al. Co-pyrolysis of waste polyester enameled wires and polyvinyl chloride: Evolved products and pyrolysis mechanism analysis. *Journal of Analytical and Applied Pyrolysis* 169, 105816 (2023).

Coralli, I., Giorgi, V., Vassura, I., Rombolà, A. G. & Fabbri, D. Secondary reactions in the analysis of microplastics by analytical pyrolysis. *Journal of Analytical and Applied Pyrolysis* 161, 105377 (2022).

R1C2: Were the authors able to find any correlations of tire wear particles with the amount of traffic in the city or observe a trend in the amount of tire wear in weekend vs weekday samples?

A-R1C2: We thank the reviewer for the valuable suggestion to investigate correlations between tire wear particles and traffic activity. Unfortunately, traffic count data for the sampling site and time of measurements were not available, which prevented a quantitative correlation analysis. The two-week sampling window also limited the robustness of potential weekday-weekend comparisons. While a slight visual variability in daily concentrations of TWPs was noted, it cannot be confidently attributed to traffic patterns without supporting data. As per the reviewer's suggestion, we attempted to perform this interpretation (Fig. a), but no clear pattern was observed in the amount of TWPs between weekend and weekday samples. In addition to this, the current study also experienced some rain events (green dots in Fig. a), which further contribute to the washout of MNPs (a separate section is added in the supplementary material; Supplementary Section S1). This is the reason we did not include this particular aspect in our main text. Although we already highlighted one insight about the number of cars and trucks registered in Saxony in 2022 (Line 194-196), which is somewhat in agreement with our results. We will also integrate continuous traffic counter data collection into an upcoming one-year monitoring campaign, enabling more robust analysis of traffic-TWP correlations in future studies.

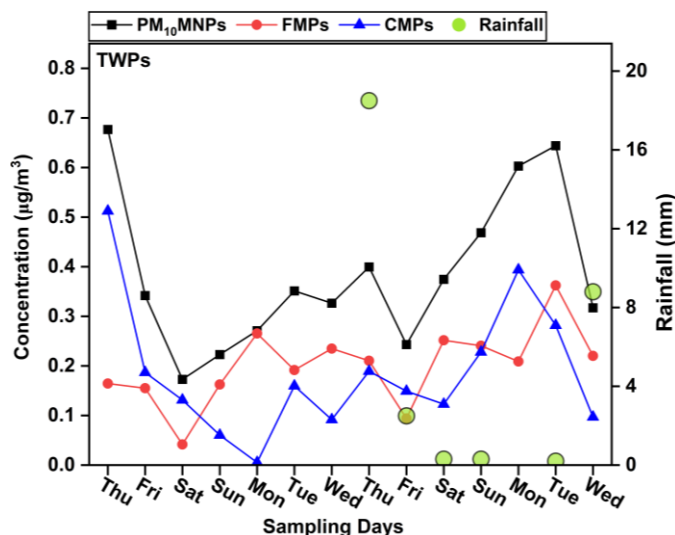


Fig a: Temporal variation of tire wear particles (TWPs) and Rain events during the sampling period.

Daily concentrations of total micro- and nano-plastics in PM₁₀ (black squares), fine microplastics (FMPs, red circles), and coarse microplastics (CMPs, blue triangles) compared with daily rainfall (green circles) during the study period.

R1C3: Line 201 – it would be helpful to the reader if the term "by mass" was added when referring to the percentages.

A-R1C3: As suggested, the term "by mass" has been added where percentage contributions are reported, to indicate that these values represent mass-based fractions.

We have amended the manuscript text as follows:

Page 12, line 232-237: In the present study, the total PM₁₀ mass load ranged from 10.9 to 24.3 µg/m³, with a mean of 16.7 ± 3.8 µg/m³. On average, 3.6% by mass of the total PM₁₀ mass was contributed by MNPs. The PM_{2.5} mass load ranged from 6.9 to 16.1 µg/m³ (mean: 11.1 ± 2.9 µg/m³), with FMPs contributing 2.8% by mass. For PM_{10-2.5}, the mass load varied between 3.4 and 8.2 µg/m³ (mean: 5.7 ± 1.5 µg/m³), with CMPs accounting for 5.2% by mass of the total mass load.

R1C4: Line 256 (and Supp Table 6) – please define OC, EC, TC, POC, SOC, POM when they are first introduced in the text.

A-R1C4: In the revised manuscript, we now provide complete definitions for all carbonaceous parameters: organic carbon (OC) (Line 319), elemental carbon (EC) (Line 326), total carbon (TC) (Line 319), primary organic carbon (POC) (Line 321), secondary organic carbon (SOC) (Line 321), and particulate organic matter (POM) (Line 330) at their first occurrence in the main text. We have also ensured that these definitions are included in the caption for Supplementary Table 6 for clarity (Results, Section 2.2.2; Supplementary Table 6 caption). Considering the comment on Supplementary Table 6, we have also updated the caption for Supplementary Table 4 for greater clarity.

R1C5: Line 423 – please provide information on what solvents were used for the dissolution of each polymer.

A-R1C5: The solvents used for dissolving the individual polymer standards during calibration are now specified in the Materials and methodology section, and a detailed overview has been included in Supplementary Table 1 for clarity and completeness.

We have amended the manuscript text as follows:

page 25, line 510-514: For calibration, individual polymer standards were dissolved and diluted in appropriate organic solvents prior to pyrolysis, including DCM used for C-PA6, C-PC, C-PS, HFIP for C-PET, acetone for C-PMMA, and a mixture of THF with DCM for C-PVC, according to their solubility¹⁰⁷ and C-PE, C-PP, CTT, TTT and MDI-PUR were weighed as solid standards (1.0-16 µg) in pyrolysis cups.

R1C6: Line 425 – a one point calibration was used for C-PP and C-PE, a method which typically cannot be reported as quantitative. I note that the cited reference Goßmann et al. explains the semi-quantitative nature of using the 1-point calibration methodology . If dissolution of the PE and PP was not possible, did the authors try to dilute in sand or another matrix when preparing calibration standards? More discussion on the limitations of a one-point calibration method is needed.

A-R1C6: We appreciate the reviewer's critical point. For C-PE and C-PP, dissolution in conventional solvents was not feasible due to their chemical characteristics. Therefore, a one-point calibration was employed, consistent with earlier approaches (Goßmann et al.,¹⁹). We did not attempt dilution in sand or other inert matrices, as this approach may introduce variability in recovery and reproducibility due to their poor homogeneity. We fully agree that the one-point calibration approach is semi-quantitative. We have expanded the material and methodology section to clarify its limitations and note that results for C-PE and C-PP should be interpreted with this constraint in mind.

We have amended the manuscript text as follows:

Page 25, Line 514-520: For C-PE and C-PP, conventional solvent dissolution was not feasible due to their non-polar, crystalline nature. Following Goßmann et al.¹⁹, we therefore employed a one-point calibration based on the lowest calibration point, which provides only semi-quantitative estimates. Alternative approaches, such as dilution in inert matrices, were considered but not applied, as they can introduce variability in recovery. Reported concentrations for C-PE and C-PP should thus be interpreted as semi-quantitative values.

R1C7: Supp Fig 1 – It would be helpful if the authors could list the equations for each calibration curve.

A-R1C7: While the calibration equations were already provided in Supplementary Table 3, we have now also added them directly to Supplementary Figure 1 to make the information immediately accessible to readers.

R1C8: Line 519 – Was a laboratory fume hood used or a laminar flow hood for the drying procedure? A typical fume hood will draw in air from the surrounding laboratory, so there would still be risk for contamination. Please comment. It would also be beneficial to the reader if the authors could provide the average amount (and standard deviation) of plastics found in the blanks, as it is noted that blank subtraction was performed (line 532).

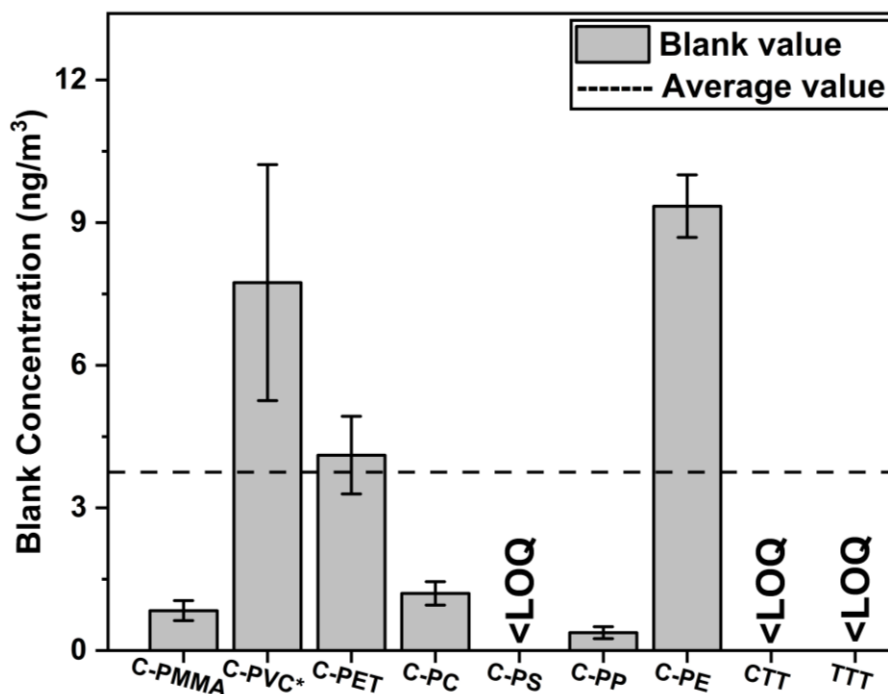
A-R1C8: We appreciate the reviewer's attention to this important point regarding contamination control. The drying procedure was conducted within a Class II biological safety cabinet (Berner Claire Pro), which utilises HEPA-filtered downward laminar airflow to protect both the samples

and the work area from airborne contamination. To quantify and correct for any residual contamination, blank filters were processed in the same manner as the samples. The mean blank concentration was 3.9 ng/m³ with a standard deviation of 0.7 ng/m³, and blank subtraction was applied accordingly in all reported results. We have now added a sentence to the Materials and Methodology section, and a supplementary figure (Figure 4) has also been included to show the average amount of plastic polymers found in the blanks.

We have amended the manuscript text as follows:

Page 30-31, Lines 640-643: The drying procedure was conducted within a Class II biological safety cabinet (Claire pro B-2-130, laminar flow hood, Berner, sample protection mode), which utilises HEPA-filtered downward laminar airflow to protect both the samples and the work area from airborne contamination.

Page 31, Line 656-660: To avoid any overestimation of the results, all sample data were raw-corrected on a raw data basis (subtraction of peak area ratios) before quantification. To illustrate the order of magnitude, we have also calculated the blank concentration of each quantified polymer. The overall average value of blanks was found to be 3.9±0.7 ng/m³ (Supplementary Figure 4).



Supplementary Figure 4: Blank concentrations of target polymers.

Blank concentrations (ng/m³) for individual polymers with error bars representing standard deviations. The dashed line indicates the average blank value across all polymers. Concentrations below the limit of quantification (LOQ) are marked as <LOQ. Polymers: polymethyl methacrylate

(C-PMMA), polyvinyl chloride (C-PVC*), polyethylene terephthalate (C-PET), polycarbonate (C-PC), polystyrene (C-PS), polypropylene (C-PP), polyethylene (C-PE), car tire tread (CTT), and truck tire tread (TTT).

R1C9: It was noted that nitrile gloves were used, however, it has been shown that the use of nitrile gloves can lead false positives in Pyrolysis GCMS experiments [Witzig et al. (2020) [<https://doi.org/10.1021/acs.est.0c03742>]]. Did the authors observe any contamination peaks in the resulting pyrograms associated with use of nitrile gloves?

A-R1C9: We thank the reviewer for bringing this important point to our attention. The study by Witzig et al. (2020) has been highly valuable in highlighting the potential for glove-derived interferences in Pyrolysis-GCMS analyses. In our work, nitrile gloves were used exclusively during the preparation of polymer standards to ensure safe handling of organic solvents, while all subsequent sample processing steps were performed without gloves to minimize the risk of contamination. To ensure no overestimation of results, we always took blank samples, and all raw data were corrected for blank values prior to quantification. The average blank concentrations ($\pm$ SD) have now been included (as described in the above comment) in the revised manuscript for transparency.

Reviewer: 2

Recommendation: Accept, with minor revisions noted.

Comments:

This paper presents the concentration of airborne micro- and nano-plastic particles over a two-week period in an urban environment in Germany. The analysis was done for PM_{10-2.5} (coarse MP; CMP) and PM_{2.5} (fine MP; FMP), along with the total MNPs. In addition, total, elemental and organic carbon were assessed from the same filters. The analytical methods are solid and represent the current state of the art for MNP (especially in the smaller size ranges). The results suggest that tire wear particles are the dominant form of airborne MNP at this urban, roadside site, followed by other common polymers: PVC, PE, PP, etc. The authors present correlation matrices to evaluate the pairwise coincidence of polymers and carbon, showing that many of the polymers are correlated with carbon. This isn't surprising, given that the MNPs are likely measured as part of the carbon (albeit as a small proportion of the total carbon), and that when particulate loads are high all types of particle abundance will increase. The more novel aspect of the study comes from the application of inhalation risk analysis, where the authors demonstrate an enhanced risk associated with exposure to urban air.

The greatest short-coming in the paper is that only one site was sampled, so any variability in the concentration of the particles is likely to come from day-to-day patterns in weather or traffic, rather than any clear variation in particle source. There was no presentation of weather variability over the study period, however, so it is unclear what caused the small variation in concentrations and relative abundance of polymers and sizes. Thus, the correlations presented don't provide as much insight as desired, since there is so little variability in the data. Perhaps a multivariate analysis (NMDS or similar) would be more helpful in illustrating patterns rather than multiple bivariate correlations. This would be a much stronger contribution if the study had been carried out at multiple sites with different potential sources, even if only at different levels of traffic or urban activities within the same city.

The manuscript is generally well written, but there are a number of places where the writing was confusing, or perhaps errors in editing. The Results/Discussion section is quite long, with a lot of detail on correlations or potential health impacts of polymers that could be removed or shortened to better streamline the results. The results and the figures are high quality and generally illustrate the patterns well. I suggest considering a different color scheme for figures 1b, 3, S3, etc because the shades of blue and green are so close that they are indistinguishable and the red/green combination may be challenging for some readers.

We thank the reviewer for their constructive overall assessment of our manuscript. We appreciate the recognition of the robustness of our analytical methods and the novelty of the inhalation risk assessment, as well as the thoughtful suggestions for improvement. We

acknowledge the limitation of sampling at a single site and have now explicitly discussed this in the Discussion section. To address variability, we have included meteorological data for the sampling period (supplementary information) and expanded the discussion on potential influences of weather and traffic patterns. In addition to the correlation analyses, we have incorporated a multivariate (NMDS) analysis to provide a more integrated view of polymer and carbon relationships. The Results/Discussion section has been streamlined to reduce redundancy, with additional details relocated to the Supplementary Information. The figures have been revised using a colour-blind-friendly palette to enhance clarity and accessibility. We believe these changes improve the clarity, focus, and overall impact of the manuscript. The above-mentioned concerns of the reviewer will be addressed through point-by-point revisions to the specific comments provided below.

Specific comments:

R2C1: Abstract

Line 21 – fine and coarse MP should be defined here.

A-R2C1: In the revised abstract, we have now defined fine microplastics (FMPs) and coarse microplastics (CMPs) at their first mention to ensure clarity for the reader.

We have amended the manuscript text as follows:

Page 1, Line 20-22: Total MNPs in PM₁₀ (PM₁₀MNPs) concentrations averaged $0.6 \pm 0.2 \mu\text{g}/\text{m}^3$, with PM_{2.5} (fine microplastics; FMPs) and PM_{10-2.5} (coarse microplastics; CMPs) contributing equally.

R2C2: Line 24 – it would help to indicate briefly which polymers were most common.

A-R2C2: We agree and have revised the abstract to indicate that the most common polymers are those that TWPs dominate, followed by PVC, PE, and PET.

We have amended the manuscript text as follows:

Page 1, Line 22-26: Tire wear particles (TWPs) were most dominant, constituting approximately 65% of total MNPs, and car tire tread particles (CTT) were consistently abundant across all studied size fractions, followed by the polyvinyl chloride (C-PVC*), polyethylene (C-PE) and polyethylene terephthalate (C-PET) clusters in the respective order.

R2C3: Line 25 – it's not clear that the data support "complex atmospheric interactions" given the lack of variability in the dataset or any investigation of mechanisms.

A-R2C3: We agree and have revised the abstract to avoid any confusion. The phrase "complex atmospheric interactions" has been replaced with "reflecting their co-occurrence in urban particulate matter," which more accurately reflects the evidence provided by our dataset.

We have amended the manuscript text as follows:

Page 1, Line 26-27: The identified polymers were strongly correlated with carbonaceous aerosol markers, reflecting their co-occurrence in urban particulate matter.

R2C4: Line 26 – the polymer-specific hazard index should be defined here, or the statement simplified to avoid confusion by the reader.

A-R2C4: We appreciate this suggestion. To improve clarity, we have simplified the sentence in the abstract by briefly defining the hazard index as a measure of relative toxicity associated with individual polymers.

We have amended the manuscript text as follows:

Page 1-2, Line 27-30: An estimated inhalation of 2.1 µg/day of airborne MNPs, combined with polymer-specific hazard index (a relative measure for polymer toxicity), may increase the relative risk of cardiopulmonary mortality by up to 9% and lung cancer-related mortality by up to 13%.

R2C5: Introduction

Line 35 – a more typical definition of microplastic is <5 mm, so limiting to <1 mm should be explained or justified using a reference.

A-R2C5: We thank the reviewer for this comment. The <1 mm definition is already justified in the manuscript with reference to Hartmann et al. (2019), who proposed this refinement in the definition.

Reference used in the main text:

Hartmann, N. B. *et al.* Are We Speaking the Same Language? Recommendations for a Definition and Categorization Framework for Plastic Debris. *Environ. Sci. Technol.* **53**, 1039–1047 (2019).

R2C6: Line 39 – this sentence mixes the matrix where MP were found with the region/location and should either be limited to region, or indicate the matrix for all regions.

A-R2C6: We agree, and to avoid confusion, we have revised the sentence so that it now refers only to the geographic regions where microplastics have been reported, without mixing in the environmental matrices.

We have amended the manuscript text as follows:

Page 2, Line 40-42: MNPs have been detected in remote geographic regions such as the North Pole³, South Pole⁴ and high-altitude sites^{5,6}, highlighting their omnipresence and long-range atmospheric transport⁷.

R2C7: Line 45 – consider rewording: "In spite of increase prevalence in the environment" and explaining what is meant by "specific single-source contribution". The latter is confusing, as MP sources are highly variable.

A-R2C7: We appreciate the reviewer's helpful comment. To keep the focus aligned with the study's objectives, we have removed the ambiguous term "specific single-source contribution" and simplified the wording. The revised sentence now emphasizes the growing environmental prevalence of airborne MNPs while acknowledging that their health and environmental impacts remain underexplored.

We have amended the manuscript text as follows:

Page 2, Line 46-48: In spite of the increased prevalence in the environment, the potential health risks associated with inhalable MNPs remain understudied.

R2C8: Line 49 - ... into the deep lung where they are suspected to cause.

A-R2C8: We thank the reviewer for bringing this to our attention. The sentence has been rephrased to improve readability and scientific precision. The revised text now clearly states the potential effects.

We have amended the manuscript text as follows:

Page 2, Line 50-53: Inhalable MNPs can be taken into the deep lung²⁶ where they are suspected of causing oxidative stress, cytotoxicity, and chronic inflammation, and potentially contributing to respiratory diseases, including interstitial lung disease and fibrosis²⁷.

R2C9: Line 51 – remove one mention of "airborne"

A-R2C9: As suggested, the use of the term "airborne" has been removed to streamline the sentence.

We have amended the manuscript text as follows:

Page 3, Line 53-56: These MNPs, once airborne, have the ability to act as carriers for co-contaminants such as heavy metals²⁸, polycyclic aromatic hydrocarbons²⁹, pharmaceuticals³⁰ and carbonaceous parameters³¹, which can amplify their toxicity.

R2C10: Line 54 – remove "There are"

A-R2C10: As suggested, the phrase "There are" has been removed to make the sentence more concise and direct.

We have amended the manuscript text as follows:

Page 3, Line 56-59: No clear regulatory thresholds for emissions and inhalation of MNPs exist, prompting the World Health Organization (WHO)³² and European Environment Agency³³ to call for standardized exposure assessments -that support their relevance to human health within the framework of Sustainable Development Goal 3 (Good Health and Well-Being).

R2C11: Line 59 – airborne MNPs are under-represented

A-R2C11: The sentence has been rephrased to improve clarity.

We have amended the manuscript text as follows:

Page 3, Line 60-63: As part of the ongoing negotiations under the Global Plastics Treaty, marine plastic pollution has now received considerable attention, but airborne MNPs are under-represented in global policy dialogues, regardless of their ubiquitous distribution and potential direct human exposure³⁴.

R2C12: Line 68 – estimates with a polymer-specific

A-R2C12: The sentence has been rephrased to improve clarity.

We have amended the manuscript text as follows:

Page 4, Line 78-81: Additionally, by integrating size-resolved exposure estimates with a polymer-specific hazard index, the present study also deduces the relative risk potential associated with MNPs inhalation, highlighting the substantial health burden posed by airborne MNPs.

R2C13: Line 88+ - Is this only true for PVC, or for other MP polymers?

A-R2C13: In our study, all polymers are reported as clusters, since pyrolysis-GC-MS produces mixed signals from homopolymers, copolymers, and polymer composites. The specific compounds associated with each cluster are provided in the Supplementary Information (Supplementary Table 3). The asterisk (*) is applied only to C-PVC to indicate its particularly complex nature, as this cluster includes contributions from PVC, other chlorinated polymers, and black carbon. To avoid confusion, we have revised the text to provide the overlapping signals of C-PVC with black carbon.

We have amended the manuscript text as follows:

Page 5, Line 101-112: To address potential overlaps with black carbon (BC), all the data related to the C-PVC cluster are labelled with an asterisk (*), which shows its mixed origin as recently shown by Goßmann et al.^{38,40}. Since thermal trace analysis of C-PVC* and related polymers only generates non-specific, aromatic indicator products, unavoidable interferences occur in complex environmental samples. In this study, Naphthalene was used as an indicator (Supplementary Table 3) for C-PVC*, despite its low specificity, due to the lack of a more selective alternative. As a result, C-PVC* is a mixed cluster with unknown proportions of chlorinated polymers on the one hand and, in particular, BC from various sources on the other. Since both PVC and BC are of high (toxicological) relevance with regard to air pollutants in the urban environment, which comes almost exclusively from anthropogenic sources^{41,42}, this cluster is both included in the overall MNPs calculations and discussed in detail.

R2C14: Line 111 – what were the weather conditions during the study and did they vary? Does this variability have any relation to the observed patterns?

A-R2C14: To address this point, we have added a description of the meteorological conditions during the two-week sampling campaign in the Supplementary Information (Supplementary Section S1). In the revised Results/Discussion section, we have added a sentence to highlight this aspect.

We have amended the manuscript text as follows:

Page 6, Line 128-130: Our observations also showed these variations, and a summary of the weather conditions during the study period is given in Supplementary Section S1.

R2C15: Line 126 – Were any statistical analyses done to evaluate the patterns, aside from the correlation analysis?

A-R2C15: A statistical analysis was indeed performed using Kernel Density Estimation (KDE) with the Kolmogorov–Smirnov test to assess the probabilistic density of MNPs intake across particle sizes. This is already presented and discussed in the manuscript (Section 3; Lines 375-388; Figure 4b).

R2C16: Line 134 – smaller than other polymers? Is it preferential, or just a characteristic of the polymer that smaller MPs are released from the original product?

A-R2C16: In the revised text, we clarify that the observed smaller size distribution of the polycarbonate cluster (C-PC) is most likely due to its inherent material properties. PC is brittle and

more prone to fragmentation under environmental stressors such as UV radiation and abrasion, leading to preferential release of smaller particles. It has to be pointed out C-PC determination based on bisphenol A dimethyl ether as an indicator ion. This is also released by epoxide-resin-based paint binders and underlines the cluster definition of the detected polymers. Surface abrasion of coatings are supposed to be of smaller sizes as well.

We have amended the manuscript text as follows:

Page 7, Line 151-155: C-PC has a unique characteristic, which appears to be entirely in FMPs, which can be attributed to its brittle nature and susceptibility to environmental degradation (e.g., UV exposure, abrasion), resulting in preferential fragmentation into smaller airborne particles^{51,52}. Additionally, binders of epoxide-based coatings are included in the C-PC. Abrasion of respective surfaces is supposed to be of a very low particle size as well.

R2C17: Line 148+ - this section reads a bit like a list of results from other studies and could synthesize these comparisons a bit more.

A-R2C17: We thank the reviewer for this constructive suggestion. In the revised manuscript, we have restructured this section to provide a more synthetic comparison rather than a list of study outcomes. We now emphasize the recurring observation that PE, PET, and tire wear particles are dominant across various urban environments, while differences in magnitude largely reflect local factors such as traffic density, waste management practices, and methodological approaches. This framing allows the section to highlight overarching patterns while still presenting relevant quantitative context from previous studies. All these changes can be seen in the answer to comment R2C18.

R2C18: Line 148 – In this section, several sentences are worded as "City reported high values". I find this wording awkward, as the cities themselves aren't reporting the values. Consider "higher levels of PE were reported for Kyoto, Japan...", etc.

A-R2C18: We appreciate the reviewer for pointing this out. We have revised the wording to ensure clarity and accuracy.

We have amended the manuscript text as follows:

Page 8, Line 168-176: Compared to the current study, Chen et al.⁴³ observed elevated levels of PVC (0.5 µg/m³) and PE (4.2 µg/m³) in PM_{2.5} at urban sites in Shanghai, China. Similarly, higher levels of PE (230 ng/m³), PET (39 ng/m³) in particle size of 0.43-2.1µm and PS (4 ng/m³) in particle size of 7.0 to 11.0µm were reported in Kyoto, Japan²¹. There was a moderate median PM_{2.5} concentrations of PET (up to 180 ng/m³), PP (up to 40.5 ng/m³), and PE (up to 33.3 ng/m³) in different classes of region observed in Graz, Austria³¹, while at high alpine sites, Kau et al.⁴⁴

reported variable PET concentrations (4.1-29.5 ng/m³ in PM₁; 0.53-35.6 ng/m³ in PM₁₀). In another urban study in China, Luo et al.²⁰ noted significantly lower PE levels (5.0-10.2 pg/m³).

R2C19: Line 156 – this statement that methodological variability limits intercomparison is very important, and a note about which methods were used for the studies cited may be warranted if there were differences in size fractions analyzed, instrumentation, extraction, etc

A-R2C19: We thank the reviewer for this helpful suggestion. We have revised the text to make clear that differences between studies can come from the choice of quantifier, instrumentation, size-segregation methods, and extraction procedures.

We have amended the manuscript text as follows:

Page 8, Line 176-179: Such differences in the concentrations most plausibly arise from methodological variability, including the choice of quantifier²⁰, instrumentation³¹, size segregation sampling²¹ and extraction procedures⁵⁷ applied for MNP analysis.

References:

Reference added to the revised manuscript:

Dierkes, G. et al. Quantification of microplastics in environmental samples via pressurized liquid extraction and pyrolysis-gas chromatography. Anal Bioanal Chem 411, 6959–6968 (2019).

R2C20: Line 174 – it may be more clear to indicate that there were two size classes and the total, since the total is the sum of the FMP and CMP

A-R2C20: We thank the reviewer for pointing out this. The sentence has been rephrased for clarity to directly state the contribution of tire wear particles (TWPs) to the measured MNP fractions. A description of PM₁₀MNPs, FMPs, and CMPs is already provided in the manuscript (Line 117-118).

We have amended the manuscript text as follows:

Page 10, Line 196: Within the present study, TWPs accounted for 60-65% in each PM size fraction

R2C21: Line 181 – remove "the" before following

A-R2C21: The suggested correction has been made in updated Line 203.

R2C22: Line 186 – this sentence is hard to follow. I'm not sure what the "indicates" is referring to

A-R2C22: As suggested, the word "indicates" is removed in updated Line 210 to make the statement more precise and easier to follow.

R2C23: Line 189 – this relationship could be evaluated statistically by doing a pairwise comparison between polymer abundance between the two size fractions and also by presenting a multivariate analysis of the two size classes to evaluate potential drivers of the overall composition of MP in each

A-R2C23: We thank the reviewer for this constructive suggestion. In response, we performed both pairwise statistical comparisons and a multivariate analysis:

1. Pairwise comparisons (Wilcoxon signed-rank tests, two-sided, n=14 pairs):
 - C-PC (p = 0.00098) and C-PMMA (p = 0.035) were significantly enriched in the fine fraction (FMPs).
 - C-PP (p = 0.041) and C-PS (p = 0.013) were significantly enriched in the coarse fraction (CMPs).
 - Other polymers showed consistent directional trends (e.g., C-PVC, TTT higher in FMPs; C-PE, C-PET higher in CMPs).
2. Multivariate analysis (Principal Component Analysis; PCA):
 - The first two principal components explained ~48% of variance.
 - FMPs clustered with C-PC, C-PMMA, C-PVC, TTT, whereas CMPs clustered with C-PE, C-PP, C-PET, C-PS.
 - This separation supports size-fraction driven compositional differences and identifies polymer groups driving the divergence.

Together, these analyses statistically and multivariately confirm our initial observation that fine and coarse microplastic fractions are compositionally distinct and dominated by different sets of polymers. We have revised the Results and Discussion accordingly and added the relevant figure and statistical summary table to the Supporting Information. In addition, we have included a clear explanation of our statistical approach (Wilcoxon signed-rank tests and PCA) in the Methods section of the revised manuscript to improve transparency. We have included a separate section for statistical analysis (section 5.5.5) in the revised manuscript.

We have amended the manuscript text as follows:

Page 10-11, Line 214-223: In order to confirm it statistically, pairwise statistical comparisons (Wilcoxon signed-rank tests, n=14) were performed, which revealed that C-PC and C-PMMA were significantly enriched in FMPs, while C-PP and C-PS were significantly enriched in CMPs. Directional trends also suggested enrichment of C-PVC and TTT in FMPs and C-PE and C-PET in CMPs (Supplementary Table 9). Principal Component Analysis (PCA) further confirmed size-fraction driven differences in polymer composition (Supplementary Figure 5). FMPs clustered

with C-PC, C-PMMA, C-PVC*, and TTT, whereas CMPs clustered with C-PE, C-PP, C-PET, and C-PS along the first two principal components (explaining ~48% of variance). This clear segregation supports our hypothesis that FMPs and CMPs are compositionally distinct, with different polymer drivers dominating each size class.

Page 29-30, Line 615-635:

5.5.5 Statistical analysis

Statistical analyses were performed to evaluate differences in polymer abundances between FMPs and CMPs. Pairwise comparisons for each polymer were conducted using the Wilcoxon signed-rank test (two-sided), a non-parametric method suitable for small sample sizes and non-normally distributed environmental data. Results are reported as medians, test statistic Wilcoxon (W), and p-values, with significance considered at $p < 0.05$. Moreover, to assess the overall compositional divergence between the two size classes and identify key polymer drivers, we performed Principal Component Analysis (PCA) on centered and standardized polymer abundance data. The first two principal components were used to visualize clustering of samples and the loadings of individual polymers.

In addition to that, in the present study, a probability density function test was performed as per Chen et al.²⁴. From the daily intake of MNPs dataset, a statistical analysis from Kernel Density Estimation (KDE) was applied to analyze MNPs exposure distributions, providing a smooth probability density function without arbitrary binning. It helped to identify dominant inhalation exposure levels across PM₁₀MNPs, FMPs, and CMPs. The Kolmogorov-Smirnov test was used to evaluate the data distribution and goodness of fit. By smoothing the data, KDE improves exposure risk characterization and supports daily intake estimates. This method enhances statistical reliability, enabling a more precise evaluation of high-risk exposure zones in urban air and strengthening the study's microplastic inhalation risk assessment. All statistical analyses were conducted in OriginPro 2023, and graphical outputs were generated accordingly.

R2C24: Figure 2 – would it be easier to see the polymers with lower abundance if the CTT and TTT were removed, or if the other polymers were blown up to better illustrate the composition of the less abundant materials?

A-R2C24: We thank the reviewer for this suggestion. In the revised figure, we have adjusted the visualization to better highlight the polymers with lower abundance.

R2C25: Line 201 – how was the total mass load measured? This isn't clear from the methods, unless I've missed something. I see the methods for the polymer analysis and the EC/OC analysis, but not total mass.

A-R2C25: We thank the reviewer for this observation. The total mass load was determined gravimetrically by weighing the filters before and after sampling under controlled laboratory conditions. To improve clarity, we have now incorporated this description into the methods section (Section 5.3) of the manuscript. We also added the measured mass load concentration in section 2.1.3.

We have amended the manuscript text as follows:

Page 27, Line 554-556: The total mass load of collected PM samples were determined gravimetrically by weighing filters before and after sampling under controlled laboratory conditions (temperature and humidity).

Page 12, Line 232-237: In the present study, the total PM₁₀ mass load ranged from 10.9 to 24.3 µg/m³, with a mean of 16.7 ± 3.8 µg/m³. On average, 3.6% by mass of the total PM₁₀ mass was contributed by MNPs. The PM_{2.5} mass load ranged from 6.9 to 16.1 µg/m³ (mean: 11.1 ± 2.9 µg/m³), with FMPs contributing 2.8% by mass. For PM_{10-2.5}, the mass load varied between 3.4 and 8.2 µg/m³ (mean: 5.7 ± 1.5 µg/m³), with CMPs accounting for 5.2% by mass of the total mass load.

R2C26: Line 201+ - this section could also be more synthetic rather than listing other studies

A-R2C26: We thank the reviewer for this valuable suggestion. In the revised manuscript, we have streamlined this section to highlight the overall patterns rather than enumerating individual study values.

We have amended the manuscript text as follows:

Page 12, Line 244-250: Despite the variability in absolute values, a clear trend emerges. MNPs, and especially TWP, consistently form a measurable part of urban particulate matter. The somewhat lower contributions observed in our study compared with locations such as Shanghai or Hamburg are likely related to differences in traffic density, sampling duration, and analytical methods. Our results fall within the range reported elsewhere and add to the evidence that MNPs are a recurring feature of urban air pollution.

Page 13, Line 253-255: The consistent presence of MNPs, together with the known toxicity of some polymer types, indicates that their health impacts may be more significant than their mass contribution.

R2C27: Line 230+ - likewise here, where this section could better synthesize the patterns, however, the lack of variability in the dataset may make further interpretation difficult, as it doesn't appear that there is much change in weather over the time period and the data are from a single source. For example, why does the widespread use of PE, PET and PS in commercial

products explain why they correlate with one another? The Wright study cited here used weather data to evaluate potential source areas of different materials, and the Chen study in Shanghai correlated MP patterns with traffic and weather data.

A-R2C27: We thank the reviewer for this insightful comment. In the revised manuscript, we have expanded the discussion of inter-polymer associations to go beyond listing correlation values and to provide clearer interpretation. Specifically, we now explain that the observed correlations among C-PE, C-PET, and C-PS likely reflect their widespread and concurrent use in packaging, consumer goods, textiles, and construction materials. Because these polymers are often released together in waste streams and share similar fragmentation and resuspension pathways, their co-occurrence in the atmosphere is expected.

We have also clarified that correlations differ by size fraction: stronger associations in fine particles may indicate shared fragmentation processes, while weaker correlations in coarse particles may reflect more complex atmospheric mixing and degradation. TWPs showed consistently strong correlations across size fractions, reinforcing their dominant role in urban MNPs emissions.

To address the reviewer's point on meteorological influences, we have explicitly acknowledged that, although meteorology can influence temporal variability in overall MNP concentrations, the relatively stable weather during our short two-week sampling period limited its role in explaining polymer-polymer correlations. Instead, the observed associations in our dataset appear to be primarily source-driven, particularly by shared usage patterns and traffic-related emissions. We refer to Supplementary Section S1 for a summary of the weather conditions during the study period.

These revisions offer a more comprehensive discussion, highlight the most plausible explanations for the observed correlations, and clearly acknowledge the scope of our dataset.

We have amended the manuscript text as follows:

Page 14, Line 281-283: Their concurrent application in packaging and consumer products, combined with similar fragmentation properties and resuspension pathways, likely explains their co-occurrence in the atmosphere.

Page 15, Line 291-293: This pattern suggests that fine fractions capture stronger source-related signals, whereas coarse fractions might be influenced by additional complex atmospheric mixing and processing.

R2C28: Line 245 – this conclusion about co-occurrence of road markings and tire particles is logical, but without evaluating sources that may vary in potential loading of these materials it is harder to draw this conclusion

A-R2C28: We thank the reviewer for this valuable remark. In the revised manuscript, we have clarified that the observed co-occurrence of road marking particles and tire wear particles is consistent with traffic-related sources, which are prominent at the sampling site. While we acknowledge that a more detailed source apportionment would strengthen this conclusion, the interpretation presented here is supported by both the site characteristics and previous studies linking these materials to traffic emissions.

We have amended the manuscript text as follows:

Page 15, Line 297-307: The observed co-occurrence of road marking particles and TWPs is consistent with traffic-related sources, which dominate at the sampling site, as also reported in earlier studies. Our observations showed meteorology-related variations, and a summary of the weather conditions during the study period is provided in Supplementary Section S1. However, because meteorological variability during the sampling window was relatively limited (Supplementary Figure 6a-d), it is unlikely to explain the inter-polymer correlations observed fully. Instead, the associations are more plausibly linked to shared usage patterns and traffic-related sources. Taken together, these findings suggest that meteorology may influence variability in overall concentrations, but polymer co-occurrence in our dataset is primarily driven by source. Therefore, further source-specific analysis would help refine this conclusion.

R2C29: Line 255 – is this statement suggesting that the carbon sum parameters are indicative of adsorbed C on the surface of the FMP? If the MP are such a small proportion of the total PM load, and also a small proportion of atmospheric particulate C, it seems that it might be challenging to detect adsorption of materials in bulk on fine relative to coarse MP particles?

A-R2C29: We agree that quantifying the adsorption of carbonaceous compounds on fine microplastics (FMPs) is challenging due to their small relative contribution to total PM and total carbon. However, several recent studies have demonstrated that even at low atmospheric abundances, fine and ultrafine MNPs can act as effective carriers for organic pollutants. For example, Rochman et al. (2013) showed significant sorption of hydrophobic organic contaminants onto plastics under field conditions, while Kirchsteiger et al. (2023) reported that ultrafine MNPs in PM_{2.5} correlated with polycyclic aromatic hydrocarbons (PAHs), suggesting a carrier role despite their small mass fraction. Similarly, Kau et al. (2024) highlighted that fine MNPs (<2.5 µm) can account for measurable contributions to organic matter and thus potentially influence adsorption-desorption dynamics.

Therefore, although MPs represent only a minor fraction of bulk particulate carbon, their physicochemical properties (e.g., large surface area-to-volume ratio, surface weathering, and polymer-specific interactions) can make adsorption processes more pronounced on fine size fractions. We have clarified this point in the revised manuscript by discussing these supporting studies.

We have amended the manuscript text as follows:

Page 15-16, Line 310-313: Although MNPs constitute only a small fraction of the total PM and carbon load, their high surface-area-to-volume ratio, particularly in the fine fraction, enables them to act as potential carriers for organic compounds, as shown in previous studies^{31,44,82}.

R2C30: Line 259 – This statement about CMP dominance needs clarification

A-R2C30: We thank the reviewer for pointing this out. Our intention was not to imply overall mass dominance, but rather to highlight that CMPs were more strongly associated with secondary organic carbon (SOC) than with primary organic carbon (POC) (Supplementary Table 6a-c). This finding suggests that CMPs may act as surfaces for condensation or adsorption of secondary organic matter formed in the atmosphere, linking them more closely with secondary processes than with direct emissions. We have revised the text to make this interpretation clearer.

We have amended the manuscript text as follows:

Page 16, Line 323-325: CMPs only observed their dominance as secondary carbon rather than primary carbon (Supplementary Table 6a-c), suggesting their potential role in interacting with secondary formation processes in the atmosphere.

R2C31: Line 263 – is this statement suggesting that PMMA, PVC, and PE are part of the POC and SOC pools? Or is there another meaning here?

A-R2C31: We thank the reviewer for this observation. Our intention here was less to focus on the fact that C-PMMA, C-PVC*, and C-PE are chemical constituents of POC or SOC, what they should be by definition. Instead, the correlation analysis (Supplementary Table 6a) shows that these polymers exhibited consistent and significant correlations with all carbon parameters ($R = 0.53$ to 0.94), including POC, SOC, and POM. This indicates that they are part of both primary and secondary organic carbon fractions, likely due to shared emission sources or parallel atmospheric processes, rather than direct inclusion in the carbon pools.

We have amended the manuscript text as follows:

Page 16, Line 328-332: In our study, only C-PMMA, C-PVC*, and C-PE within PM₁₀ MNPs showed strong correlations with all carbon parameters ($R = 0.53-0.94$), demonstrating their strong association with particulate organic matter (POM) and indicating that they occur as particle-

bound species embedded in organic matrices while coexisting within both primary (POC) and secondary (SOC) carbonaceous fractions (Supplementary Table 6a).

R2C32: Line 285 – I'm not certain that the data support this statement entirely. The correlations are based on a single site, and any variability may be due to simple shifts in traffic or wind that kick up particles, so that the abundance of all particles in the atmosphere will increase concomitantly. Is there evidence that the MPs are aggregated directly with or adsorbed to the carbonaceous particles? Or just that they co-occur?

A-R2C32: We thank the reviewer for this thoughtful comment. While the correlations observed between MNPs and carbon parameters do suggest co-occurrence, we acknowledge that adsorption of organic carbon to microplastic surfaces, including carbonaceous material, is a known phenomenon. For instance, Rochman et al. (2013) demonstrated that microplastics can adsorb hydrophobic organic contaminants, supporting the idea that fine MNPs might similarly adsorb carbonaceous particles under certain conditions. However, due to the small size and low mass fraction of MNPs in our dataset, direct adsorption remains challenging to conclusively establish based on our current findings.

Thus, while we propose that the correlations reflect co-occurrence due to common sources or transport processes, we also acknowledge the possibility that MNPs, especially the finer fractions, may be adsorbing organic carbon from the surrounding atmosphere, as suggested by the literature. We have updated the manuscript to reflect this perspective. We have amended the manuscript text as follows:

Page 17, Line 351-358: The observed correlations between MNPs and carbon fractions suggest co-occurrence in the aerosol phase, likely driven by common emission sources or atmospheric transport processes. Although the small mass fraction of MNPs in this study makes direct adsorption difficult to establish, previous studies⁸² have documented the ability of microplastics to adsorb carbonaceous material. Therefore, it remains plausible that fine MNPs in our samples also adsorbed organic carbon from the surrounding environment, contributing to the observed correlations.

R2C33: Line 296+ - this section had a number of sentences that were hard to follow, and perhaps were editing errors, especially line 304, 307, 310, 311

A-R2C33: We thank the reviewer for highlighting this section. We have revised the sentences for better clarity and flow, particularly in Lines 304, 307, 310, and 311. The revised text now clearly presents the daily and annual intake estimates, as well as the comparison to other studies, ensuring that the intended meaning is easily understood.

We have amended the manuscript text as follows:

Page 18, Line 360-363: Inhalation of airborne MNPs is increasingly recognized as a potential exposure pathway, though their health impacts are not fully understood. According to the WHO, mass concentration is a crucial parameter for evaluating air pollution exposure and its health effects, along with developing legislation to address public exposure to MNPs⁸⁵.

Page 18, Line 369-379: The study estimated a daily inhalation exposure of 2.1 µg of PM₁₀MNPs, including 1.1 µg of FMPs and 0.94 µg of CMPs (Figure 4a). Upscaling these inhalation exposure estimates to annual intakes of 0.7 mg, 0.4 mg, and 0.3 mg, respectively, is essential for evaluating long-term health risks and ensuring that laboratory findings reflect real-world exposure for meaningful dose-response assessments. These annual intakes are comparable to those reported in an Australian indoor house dust study in Sydney⁸⁶ (0.2 mg/kg-body weight/year). An additional study in Taiyuan, China²⁰, estimated the daily intake of PE bound to PM_{2.5} at 34 pg/day. In contrast, a broader range of MNPs intake, from 61.85 to 102.59 µg/day, was observed in Mumbai⁸⁷, India. However, as the analytical methods used in these studies are neither standardized nor consistent, evaluating these differences remains challenging, as highlighted in Section 2.1.1.

R2C34: Line 304 – this statement seems to be mixing routes of exposure between airborne and food-based sources

A-R2C34: We thank the reviewer for pointing this out. To avoid confusion between the different exposure routes, we have removed the sentence that mixed airborne and food-based MNP exposures.

R2C35: Line 313 – it isn't clear which findings are referred to here – the present study or the Mumbai study?

A-R2C35: We thank the reviewer for this observation. To improve clarity, we have revised the text.

We have amended the manuscript text as follows:

Page 19, Line 381-384: Based on the modelling approach findings, it appears that airborne MNPs intake may be higher in the present than previously estimated, emphasizing the value of analytical measurements to complement model-based exposure assessments.

R2C36: Line 340+ - the hazard categories (line 342), associated chemicals (lines 347+) and potential human health impacts (lines 353, 355, 356, etc) could be put into a table that summarizes the potential toxicity associated with prior studies, and this section streamlined to focus more on the data in hand and which materials may pose the greatest risk

A-R2C36: We thank the reviewer for this helpful suggestion. To enhance clarity and focus on the data from our study, we have summarized the hazard categories, associated chemicals, and potential human health impacts into a summary table. This allows for a clearer comparison of toxicity across the materials studied while streamlining the discussion to emphasize the materials with the greatest potential risk. We have revised the manuscript accordingly to focus more on the data in hand and highlight which materials pose the greatest risk.

We have amended the manuscript text as follows:

Page 21, Line 438-441: To improve clarity and focus on the key findings, we have consolidated the hazard categories, associated chemicals, and potential human health impacts into Supplementary Table 7, facilitating a direct comparison of toxicity data

R2C37: Line 383 – space after comma

A-R2C37: We thank the reviewer for this minor but important observation. The requested space has been added.

R2C38: Line 405 – the sentences here appear incomplete

A-R2C38: We thank the reviewer for pointing this out. The sentence has been revised to improve clarity and completeness.

We have amended the manuscript text as follows:

Page 23-24, Line 479-483: Commercially available raw polymers were used as standards to quantify MNPs in air. A total of eleven polymers were selected, including PE, PP, PVC, PET, PS, PMMA, PC, PA6, MDI-PUR, CTT, and TTT. Detailed information on these polymer standards used for identification and quantification of the respective clusters including suppliers, is provided in Supplementary Table 1.

R2C39: Line 410 – were filtered

A-R2C39: The sentence has been updated to reflect the correct verb tense.

We have amended the manuscript text as follows:

Page 24, Line 487-489: All solvents were filtered through a glass fibre filter of 0.3 µm pore size (Whatman, Altmann Analytical, Germany; pretreated at 500°C/4h).

R2C40: Line 430 – check capitalization

A-R2C40: The capitalization has been reviewed and corrected in the manuscript.

R2C41: Line 442+ - I believe that the PM_{2.5} and the PM₁₀ were measured separately on the two different instruments deployed in the field, but this could be made more clear

A-R2C41: We thank the reviewer for this helpful comment. We have clarified in the manuscript that PM_{2.5} and PM₁₀ were indeed measured separately using different instruments during the field campaign. This distinction is now explicitly stated in the Methods section.

We have amended the manuscript text as follows:

Page 26, Line 536-538: Particulate matter (PM) samples (PM₁₀ and PM_{2.5} fractions simultaneously) were collected at Torgauer Straße, Leipzig, Germany (51.35° N, 12.42° E) using two separate DIGITEL (DHA-80) high-volume samplers (Supplementary Figure 2).

R2C42: Line 506 – by background levels, do you mean the levels of PM without microplastic?

A-R2C42: That is correct, in this context, "background levels" refers to the concentration of PM_{2.5} that would exist in the absence of anthropogenic emissions, including microplastics.

We have amended the manuscript text as follows:

Page 29, Line 605-606: which refers to the concentration of PM_{2.5} that would exist in the absence of anthropogenic emissions, including microplastics.

Reviewer: 3

Recommendation: Minor revisions.

Comments:

This work provides a comprehensive analysis of airborne micro- and nanoplastics (MNPs) in urban air, highlighting tire wear particles (TWPs) as the dominant source and quantifying their potential health risks through inhalation exposure. The use of Py-GC-MS for polymer-specific characterization is robust, and the integration of carbonaceous aerosol interactions adds valuable insights. However, limitations include methodological ambiguities (e.g., C-PVC* cluster interpretation), insufficient discussion of comparative studies, and overextrapolation of health risks without polymer-specific toxicological validation.

We sincerely thank reviewer 3 for their positive view, thorough evaluation of our work and for the constructive comments that will further strengthen the manuscript. We greatly value the positive recognition of the robustness of our polymer-specific characterization using Py-GC-MS and of the additional insights gained from integrating carbonaceous aerosol interactions. In response to the reviewer's suggestions, we have revised the manuscript to provide greater methodological clarity, expand the comparative context with previous studies, refine the framing of the health risk assessment, and add further justification for our experimental design. A detailed point-by-point response is provided below.

R3C1: Clarify how C-PVC* cluster differentiation from black carbon (BC) was handled, given its mixed signals (Page 4-5). Statistical methods for correlation analysis (e.g., p-values) are missing in Supplementary Table 6.

A-R3C1: We thank the reviewer for this valuable comment. The complexity of the C-PVC* cluster and its overlap with black carbon (BC) has already been addressed in the manuscript (page 5, lines 101-103). All C-PVC data are marked with an asterisk (*) to indicate this mixed origin, consistent with previous work (Goßmann et al.,^{38, 40}). As detailed in the text, thermal trace analysis produces mainly non-specific aromatic indicator products, and Naphthalene was selected as a pragmatic though non-exclusive marker (Supplementary Table 3). Thus, C-PVC* represents a combination of chlorinated polymers and black carbon, both of which are toxicologically relevant, and for this reason, the cluster is included in the overall MNPs calculation and discussed in detail. We have rephrased our explanation in the main text for more clarification.

In addition, we have included the p-values in Supplementary Table 6 and clarified in the main text that correlations were considered significant at $p < 0.05$.

We have amended the manuscript text as follows:

Page 5, Line 101-112: To address potential overlaps with black carbon (BC), all the data related to the C-PVC cluster are labelled with an asterisk (*), which shows its mixed origin as shown recently

by Goßmann et al.^{38,40}. Since thermal trace analysis of C-PVC* and related polymers only generates non-specific, aromatic indicator products, unavoidable interferences occur in complex environmental samples. In this study, Naphthalene was used as an indicator (Supplementary Table 3) for C-PVC*, despite its low specificity, due to the lack of a more selective alternative. As a result, C-PVC* is a mixed cluster with unknown proportions of chlorinated polymers on the one hand and, in particular, BC from various sources on the other. Since both PVC and BC are of high (toxicological) relevance with regard to air pollutants in the urban environment, which comes almost exclusively from anthropogenic sources^{41,42}, this cluster is both included in the overall MNPs calculations and discussed in detail.

In addition to this, due to the frequent use of the black carbon word in the text, we have now abbreviated this to BC. The reviewer can find this change in lines 102, 109, 347, and 432.

Page 14, Line 270-272: Pearson's correlation coefficient (R) was calculated (Supplementary Table 6a to c) with p value <0.05 to check the interaction between the detected polymers and carbon sum parameters in their respective size fractions.

References:

Reference added to the revised manuscript:

Niranjan, R. & Thakur, A. K. The Toxicological Mechanisms of Environmental Soot (Black Carbon) and Carbon Black: Focus on Oxidative Stress and Inflammatory Pathways. *Front. Immunol.* **8**, 763 (2017).

R3C2: Discrepancies in MNP concentrations (e.g., Shanghai: 5.6 µg/m³ vs. Leipzig: 0.6 µg/m³) lack discussion on methodological differences (sampling, analysis) or emission source variations (Page 6).

A-R3C2: We thank the reviewer for this observation. The discrepancies in reported MNP concentrations between sites (e.g., Shanghai vs. Leipzig) are mainly due to the use of different quantifiers for measurement, which makes direct comparisons difficult. We have clarified this point in the revised manuscript.

We have amended the manuscript text as follows:

Page 7, Line 139-143: It should be noted that discrepancies in reported MNP concentrations across sites are partly due to differences in quantification strategies, particularly the choice of marker compounds/quantifiers, which limits the direct comparability of absolute values in addition to variations in local emission sources.

R3C3: Extrapolating PM_{2.5} risk models to MNPs requires justification, as polymer-specific toxicity (e.g., C-PVC* PHI) may not align with generic PM toxicity (Page 18-19).

A-R3C3: We appreciate the reviewer's insightful comment. We agree that extrapolating PM_{2.5} risk models to MNPs has limitations, as polymer-specific toxicities may differ from generic PM. In our study, the PM_{2.5} framework is applied only as a first-order approximation in the absence of comprehensive MNP-specific dose–response data. To account for polymer-specific hazards, we calculated the Polymer Hazard Index (PHI) following the approach of Lithner et al. (2011), which assigns toxicity scores based on monomer hazard classifications. This combined strategy allows us to frame potential health risks while highlighting the urgent need for more refined, polymer-specific toxicological datasets. We have revised the text (Page 18) to make this justification explicit.

We have amended the manuscript text as follows:

Page 20, Line 411-419: To incorporate polymer-specific considerations, we calculated the Polymer Hazard Index (PHI) according to Lithner et al.⁴², which is based on monomer hazard classifications. This approach, combined with PM_{2.5} risk models, serves as a first-order approximation in the absence of polymer-specific dose-response data, while underscoring the need for further toxicological research on MNPs. Polymers like C-PE, C-PET, and C-PP have been categorized as IV, III, and I, respectively, with PHI shown in Supplementary Table 7 indicating C-PE is more hazardous compared to the other polymers (C-PET and C-PP) and can cross the blood-brain barrier, potentially leading to neurotoxic effects and neurodegenerative diseases^{96,97}.

R3C4: Justify why Torgauer Straße (high traffic) was chosen as the sole sampling site. Urban background vs. roadside variability should be addressed (Page 23).

A-R3C4: We agree that the chosen site represents a roadside location; however, as Torgauer Straße lies within the urban area, it also reflects urban exposure conditions, serving as an upper-limit scenario due to its high traffic density. A minor amendment has been made to the text (Page 24) to clarify this rationale, and we further acknowledge the reviewer's suggestion by noting that future investigations should include multiple sites, encompassing both roadside and urban background locations, to capture the variability of MNP concentrations across urban environments.

We have amended the manuscript text as follows:

Page 26, Line 538-548: The roadside site, located within the urban area, represents an upper-limit exposure scenario due to its high traffic density. Sampling was conducted over a two-week period (01 to 14 September 2022), which provides a focused and detailed snapshot of MNPs composition under conditions of strong traffic influence. This setup offers the advantage of capturing peak urban exposure levels at fine size resolution of PM and generating high-quality baseline data for health risk estimation. At the same time, the restricted spatial and temporal scope limits the

representativeness of our findings for other environments (e.g., suburban or rural) and prevents assessment of seasonal or long-term variability. A more representative picture could be obtained in future research by including diverse sites (urban and rural background) and conducting longer-term sampling to reflect broader variability in MNPs concentrations.

R3C5: One-point calibration for C-PE/C-PP (low concentrations) is inadequate. Provide detection limits and error margins (Page 22).

A-R3C5: We thank the reviewer for highlighting this point. For C-PE and C-PP, conventional solvent dissolution was not feasible due to their non-polar and crystalline character. As described previously (Goßmann et al.¹⁹), we therefore applied a one-point calibration, which allows only semi-quantitative estimation at low concentrations. Limit of detection (LOD) and Limit of quantification (LOQ) have now been provided in the revised manuscript (Supplementary Table 3) to improve transparency. Accordingly, reported concentrations for C-PE and C-PP should be regarded as semi-quantitative, and we highlighted this aspect in the revised manuscript.

We have amended the manuscript text as follows:

Page 25, Line 514-520: For C-PE and C-PP, conventional solvent dissolution was not feasible due to their non-polar, crystalline nature. Following Goßmann et al.¹⁹, we therefore employed a one-point calibration based on the lowest calibration point, which provides only semi-quantitative estimates. Alternative approaches, such as dilution in inert matrices, were considered but not applied, as they can introduce variability in recovery. Reported concentrations for C-PE and C-PP should thus be interpreted as semi-quantitative values.

R3C6: Excluding C-PA6/MDI-PUR due to "undetectable signals" (Page 5) may bias results. Discuss potential underestimation of total MNP mass.

A-R3C6: We acknowledge that excluding C-PA6 and MDI-PUR due to undetectable signals may contribute to a potential underestimation of total MNP mass. This outcome is likely related to the relatively short sampling duration applied in this study. To address this, a clarification has been added to the revised text (Page 5).

We have amended the manuscript text as follows:

Page 5-6, line 112-116: This study does not quantify C-PA6 and MDI-PUR due to undetectable signals (<LOD) in the analyzed samples, as their contribution is supposed to be very low, which may contribute to a potential underestimation of total MNP mass. This limitation is likely related to the relatively short sampling duration; therefore, a longer sampling duration is required to enhance detection sensitivity for such polymers.

R3C7: Daily intake calculation assumes fixed outdoor exposure time (4h). Validate with local activity patterns or sensitivity analysis (Page 24).

A-R3C7: In the present study, the daily intake calculation was carried out using a standard exposure model, assuming an inhalation rate of 0.83 m³/h and a fixed outdoor exposure time of 4 hours/day. This assumption follows WHO and USEPA guidance and is consistent with recent MNP inhalation studies (e.g., Luo et al., 2023). While detailed time-activity data were not available for this study, we have clarified this limitation in the revised text (Page 26) and noted that future work should incorporate local activity surveys or sensitivity analyses to capture variability in exposure better.

We have amended the manuscript text as follows:

Page 28, Line 589-591: However, as detailed time-activity data were not available for this study, future assessments should incorporate local activity surveys or sensitivity analyses to capture variability in exposure better.

R3C8: SDG linkages (Page 20) are generic. Specify actionable policies (e.g., tire wear regulations, MNP monitoring standards).

A-R3C8: We agree that the initial discussion of SDG linkages was presented in a more general manner. To address this, we have revised the text (Page 23) to include concrete policy examples, such as the regulation of tire wear particles, the establishment of standardized MNPs monitoring protocols, and the integration of microplastic indicators into urban air quality management frameworks. These examples strengthen the connection between our findings and actionable policy measures that align with relevant SDGs.

We have amended the manuscript text as follows:

Page 23, Line 469-471: In this context, actionable measures include regulating tire wear emissions, establishing standardized monitoring protocols for airborne MNPs, and integrating polymer-specific indicators into existing air quality regulations.

R3C9: Abbreviations (e.g., FMPs/CMPs/TWPs) are defined multiple times (Abstract, Section 2.1.1). Consolidate definitions in the Introduction.

A-R3C9: We agree, and to improve clarity and avoid redundancy, we have consolidated all abbreviations in the Introduction, where they are defined once and then consistently used throughout the manuscript. Minor repetitions in the Abstract were retained only where essential for readability.

We have amended the manuscript text as follows:

Page 4, Line 73-75: Within the present study, we address the aforementioned uncertainties by applying Py-GC-MS for detailed polymer-specific characterization of size-segregated airborne MNPs in PM₁₀ (PM₁₀MNPs), PM_{2.5} (Fine microplastics; FMPs) and PM_{10-2.5} (Coarse microplastics; CMPs).

Page 6, Line 117-118: In the present study, concentrations of MNPs in PM₁₀MNPs, FMPs and CMPs are reported, where CMPs were calculated by subtracting the concentration of FMPs from PM₁₀MNPs.

R3C10: Contrast Py-GC-MS with other techniques (e.g., μ FTIR, Raman) in Introduction/Discussion to highlight methodological advantages.

A-R3C10: We thank the reviewer for this constructive suggestion. In line with the comment, we have added a paragraph in the Introduction (Page 3) contrasting Py-GC-MS with other commonly used techniques such as μ -Raman and μ -FTIR. The revised text highlights the strengths and limitations of these approaches and explains why Py-GC-MS was selected as particularly suitable for complex environmental matrices such as airborne MNPs.

We have amended the manuscript text as follows:

Page 3, Line 64-72: There are several techniques available for analyzing airborne MNPs, including vibrational spectroscopy methods such as μ -Raman³⁵ and μ -FTIR³⁶, which provide information on particle size, number, and polymer composition. However, their detection limits ($\geq 1 \mu\text{m}$ for μ -Raman, $\geq 10 \mu\text{m}$ for μ -FTIR) and susceptibility to fluorescence or organic matter interference restrict their applicability, particularly for nano-plastics. The pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS) approach, on the other hand, provides polymer-resolved, mass-based quantification with high sensitivity and comparable less pretreatment. Thus, Py-GC-MS is particularly suitable for samples in complex environments such as airborne MNPs.

References:

Reference added to the revised manuscript:

Trainic, M. *et al.* Airborne microplastic particles detected in the remote marine atmosphere. *Commun Earth Environ* **1**, 64 (2020).

Chen, Q. *et al.* Long-range atmospheric transport of microplastics across the southern hemisphere. *Nat Commun* **14**, 7898 (2023).