

We would like to thank the referee for the effort and time spent reading our manuscript and posing questions and comments, which improve its relevance and clarity. Please find our responses in the following document. All comments are individually replied to.

Review:

“Is transport of microplastics different from that of mineral dust? Results from idealized wind tunnel studies”

Summary:

In their manuscript, Esders and colleagues investigated the detachment process of spherical polyethylene (diameters: 42, 69, 115 μm) and borosilicate (diameter: 69 μm) particles from a glass slide surface using wind tunnel experiments. The study explored the influence of the substrate's hydrophobicity/hydrophilicity on the detachment process of the particles. The authors employed different cleaning and coating strategies to coat the substrates accordingly. In addition, they investigated the impact of relative humidity on the detachment process. They didn't directly control the relative humidity but rather used filtered air connected to the outside air source, with humidity levels measured continuously throughout the experiments, varying between 20 and 60 %. Furthermore, the researchers included the examination of impact-induced detachment from the spheres themselves in their data set. By comparing their experimental data with a model, the study ensures validation of their findings.

The primary aim of this explorative study is to gain first insights into the release mechanisms of microplastics in the atmosphere, starting with the investigation of one type of microplastic and comparing it to a well-studied mineral particle. The manuscript makes a valuable contribution to the field of microplastic aerosol research. Considering the significance of its findings, the work presented certainly warrants publication in ACP, after major revisions.

GENERAL COMMENTS:

- One major concern I have with the manuscript is the lack of discussion on the diversity of microplastics (MP). The authors exclusively studied one type of microplastic, namely polyethylene, without acknowledging the vast diversity that exists within the category of MP. Even within polyethylene (PE), there are different variations like high density PE (HDPE), linear low-density PE (LLDPE), etc. each with varying shapes, additives, and characteristics. Consequently, the title may be misleading, as the conclusions drawn are limited in scope and pertain solely to the spherical PE particles utilized in the study, not all microplastics.

Additionally, the manuscript does not address the fluorescent nature of the PE particles used. I assume that the PE particles contain a dye that fluoresces. In this case, could the dye have an influence on the results? If yes, are the findings then comparable to other spherical PE particles that lack the fluorescent dye? I think it would be beneficial if the authors took measures to validate the material.

The material is 30 % fluorophore, trade secret of cospheric, but it is evenly dispersed (answer from the Company), thus we can assume, that the surface is influenced 30% fluorophore.

Further, the leakage of dyes from particles is a process that could influence the surface chemistry. However, to the best of our knowledge there is only a number of studies that show that this process could represent a problem in mild aqueous solutions and certainly not on the time scales considered here (e.g. Kodger, Thomas E. u.a. (2017): Stable, Fluorescent Polymethylmethacrylate Particles for the Long-Term Observation of Slow

Colloidal Dynamics, in: Langmuir 33(25), 6382-6389. DOI: 10.1021/acs.langmuir.7b00852.). As the process takes place in air, the presence and contamination by volatile organic components will be much more dominating. However, the latter process would be present for all samples in equal manner and cannot be suppressed within the experimental setup.

For instance, conducting Raman or FTIR spectroscopy could provide insights into the presence and distribution of the dye or any other additives on the surface of the plastic particles used. The authors show in their manuscript that the surface chemistry of the substrate influences the detachment process of the particles, so I assume the surface chemistry of the particles is also important, right?

The reviewer mentions an important point that has been admittedly not addressed sufficiently in the manuscript: The surface chemistry of both, the particles, and substrates, are of importance. However, not due to the specific functional groups but due their wetting behavior (i.e. hydrophilic vs. hydrophobic) that will control the formation of the capillary bridge between the particle and substrate (Butt, Hans-Juergen (2008): Capillary forces: Influence of roughness and heterogeneity 24(9), 4715-4721. DOI: 10.1021/la703640f.). Only in the case of silica and amino-modified slides the formation of chemical bonds in the contact area can be expected. The most promising technique to elucidate capillary and chemical forces would be the so-called colloidal probe technique, where a micro-meter sized particle of analogous composition is attached to an AFM-cantilever and the force required to remove the particle from the substrate can be measured with a force resolution in the order of a few pico-Newtons (Kappl, Michael/Butt, Hans-Jürgen (2002): The colloidal probe technique and its application to adhesion force measurements, in: Particle & Particle Systems Characterization: Measurement and Description of Particle Properties and Behavior in Powders and Other Disperse Systems 19(3), 129-143. DOI: 10.1002/1521-4117(200207)19.; Leite, FL/Herrmann, PSP (2005): Application of atomic force spectroscopy (AFS) to studies of adhesion phenomena: a review, in: Journal Of Adhesion Science And Technology 19(3-5), 365-405. DOI: 10.1163/1568561054352667.). However, this technique is unfortunately out of the scope for this study.

Additional Comment:

We changed the plot showing the ratio of the idealized fluid threshold (previous CIMs) and collision threshold (previous CDMs) as a function of the idealized fluid threshold. Plotting any mathematical operation of A and B as a function of A leads to spurious correlation. Thus, we changed to plotting the collision thresholds as a function of the idealized fluid thresholds. And describe the difference between them, as the result of collisions. See Section 3.1.

SPECIFIC COMMENTS.

Title:

- The title appears to be misleading as it seemingly generalizes the study to all MP, while the research focuses solely on spherical PE particles.

We changed the title of the manuscript to: Is transport of microplastics different from mineral particles? Idealized wind tunnel studies on polyethylene microspheres. Thus, it should be clear, that we focus on one prominent representative of microplastics.

Introduction:

- In general, the Introduction is well-composed, encompassing the current state of the art and adequately essential aspects.

Thanks for the positive feedback.

- Page 2; Line 42: I miss the following citations: Materić, et al., 2022

We added the citation. Line 46

- Page 2; Line 46, 47: There are multiple detachment processes that can occur in the environment through which MP can be released into the atmosphere. For instance, one mechanism involves MP release through bubble bursting in the ocean, which differs from the phenomena that govern detachment from soil. To provide clarity, specify and explain the detachment processes investigated by Tian et al. (2022) and Yang et al. (2022) in the second sentences of this paragraph.

We added a description of the emission mechanisms.

Methods:

- All equations miss numbering.

We added numbering to all equations.

- Page 4; Equation 1: “u” is not defined in the text.

We added a definition of u.

- Page 5; Line 115: Figure A5 is wrongly linked here.

We corrected the link.

- Page 5; Equation 2: “κ” is not defined in the text.

We added a definition of κ.

- Page 5, Line 121: The authors could perhaps briefly explain how the algorithm works to facilitate understanding of the methods.

We added a brief explanation.

- Page 5; Line 124: Can you add the step size?

The step size varied, as the adhesive forces varied, due to size, material and air humidity. Thus, we choose to describe the step-size with the intention of a similar percentage of detached microspheres for each step.

- Page 5, Chapter 2.4: The authors do not discuss why they chose PE particles for the study. Is there any specific reason for it?

We used these specific PE microspheres, as PE is one of the commonly used plastics, it was readily available as fluorescent microspheres, that are necessary for a robust detection, and borosilicate microspheres with the same diameter were available as reference material.

- Page 6; Table 1: Why is the contact angle for “substrate a” not described as a discrete number?

For substrate a no discrete contact angle could be determined, because water completely wets the substrate and no droplet forms, indicating a highly hydrophilic substrate.

- Page 6; Line 134-136: Which method (image processing technique etc.) did the authors use to quantify the roughness of the particles? Also, the link to the Figures in the Appendix is missing.

We added the link to the appendix. The method is described in lines 395-406

- Page 6; Line 142: Please elaborate in more detail how the microspheres were deposited onto the substrates. Gravitational settling is not a sufficient explanation.

We added further explanation, on how the microspheres were deposited. See lines 163-165.

- Page 15, Chapter A2: I miss the discussion on how the hydrophilicity changes by the cleaning procedures. Which reactions are occurring that the surface energy changes?

The cleaning procedure applied is well known in the field of surface science and goes back to Kern and coworkers.[Kern, W/Puotinen, D A (1970): Cleaning Solutions Based on Hydrogen Peroxide for Use in Silicon Semiconductor Technology, in: RCA Review 31(2), 187.] It is known under the name ‘RCA-cleaning’, after the company where it has been developed. Here, only the second part of the cleaning procedure has been utilized, which comprises a controlled oxidation of the surface to form Si-OH groups at the surface.

Results & Discussion:

- Page 11; Figure 4: The caption claims that the data in the graph represents box plots. However, it is evident that it is in fact not. Please provide the correct representation or update the caption accordingly for clarity.

We corrected the caption.

- In general, the authors have chosen to name the substrates alphabetically, referring to them as "substrate a," "substrate b," etc. However, this can be somewhat overwhelming when seen independently in the text. To improve readability and clarity, I would recommend using italics or

capital letters to distinguish the substrates, such as "Substrate A," "Substrate B," and so on. This will make it easier for readers to identify and follow the different substrates throughout the text.

We changed to the proposed representation.

- Page 14; Chapter 3.4 (Figure 9): Chapter 3.4 misses the discussion regarding the comparison of data of PE115 & PE42 from Figure 9 to PE69, as shown in Figure 5.

We added discussion regarding PE115 and PE42 in the context with PE69 and GL69. See lines 315-318.

Conclusion:

- As the dominant fraction of MP found in our environment are rather irregular-shaped fragments and fibres, wouldn't the authors think it would be a good reason to study these as well as they are environmentally more relevant than spheres?

We agree that studying MP geometries that occur more often in the environment are important. Studying fibers and irregularly shaped particles should be a priority in the future. However, this manuscript presents a first step in studying the atmospheric transport of MP, with a focus on simple geometry and idealized substrates. In the future we will use the insights from the current manuscript to work on more complicated particles, substrates, and emission mechanisms in a controllable environment.

Appendix A:

Page 17; Line 333: There is a space missing between the sentences: [...] (see Fig.A4) At [...]"

We corrected the spacing.

References:

Materić, D., Kjær, H. A., Vallelonga, P., Tison, J. L., Röckmann, T., & Holzinger, R. (2022). Nanoplastics measurements in Northern and Southern polar ice. *Environmental research*, 208, 112741.

Tian, X., Yang, M., Guo, Z., Chang, C., Li, J., Guo, Z., Wang, R., Li, Q., & Zou, X (2022).: Plastic mulch film induced soil microplastic enrichment and its impact on wind-blown sand and dust, *Science of The Total Environment*, 813, 152 490

Yang, M., Tian, X., Guo, Z., Chang, C., Li, J., Guo, Z., Li, H., Liu, R., Wang, R., Li, Q., & Zou, X. (2022): Effect of Dry Soil Aggregate Size on Microplastic Distribution and Its Implications for Microplastic Emissions Induced by Wind Erosion, *Environmental Science & Technology Letters*, 9, 618–624