RC2: 'Comment on soil-2021-136', Frederick Büks, 05 Jan 2022 reply

In their present work, the authors analyse the alteration of soil structural, soil hydrological and erosion parameters after addition of plastic microfibers. In the first of two experiments, disaggregated soil samples from three locations with different texture (Lt3, Ls2 and Slu, following KA5) were incubated for 6 month to compare the re-formation of total and water-stable aggregates, macroporosity/air capacity and plant available water capacity between treatments with and without microplastic (MP). In a second experiment with similar preparations, the erodibility of soil samples was tested in runoff containers by measuring surface water runoff, drainage and sediment loss. These parameters are known to be influenced by a loss of soil structure and water holding capacity. The combination of both experiments is therfore a good approach to analyse cross-scale effects of MP contamination in soils. Furthermore, MP fibers are a major part of plastic brought into agricultural fields by the application of sewage sludge and have been shown to be adverse to soil structure. The applied concentration of 0.5 % w/w only apperas in highly contaminated areas, but is reasonable. On the other hand, the authors used juvenile MP, which has surface characteristics very different from weathered MP found in the environment. This work can make a significant contribution to our knowledge of the effects of MP on soil structure and landscape erosion. However, from my point of view some improvement is needed. In the following I would like to list a few points which I hope will be helpful for you.

Authors (ACs): We would like to thank Dr. Frederick Büks for the thoughtful comments and suggestions which increased the clarity and the quality of our manuscript

Abstract

Line 20: Please describe in more detail.

ACs: We modified the abstract according to the Referee’s advice

Introduction

Line 28: „Improper disposal“ sounds a little bit like littering, but MP in remote areas is more from textile fibers or intended application followed by dispersal.

ACs: We changed “*Improper disposal*” to “*Mishandling*”

Line 35: Is it „sewage sludge“ (without comma) or do you mean „sewage, sludge“ in the sense of „waste water and sewage sludge“? There are also other entry pathways like coatings of seeds and fertilizers (very important) as well as irrigation from natural water bodies.

ACs: We modified the sentence according to the Referee’s suggestion

Line 38: Slow degradation – please add a reference.

ACs: We added the following reference in the revised manuscript: O'Kelly, et al. 2021 (O'Kelly, B.C., El-Zein, A., Liu, X., Patel, A., Fei, X., Sharma, S., ... & Singh, D.N. (2021). Microplastics in soils: An environmental geotechnics perspective. Environmental Geotechnics, 40(XXXX), 1-33)

Line 42: … percolation, cryoturbation, peloturbation.
ACs: We included "*cryoturbation etc.*" in the revised text

Line 46: They are related, cancel „more or less strictly".
ACs: We modified the text according to the Reviewer's comment

Line 56: This work is more on changed soil properties than the underlying processes.
ACs: We modified the text according to the Reviewer's comment

Lines 58-59: Is the expected impact assumed to be positive or adverse? Why do you assume that the fragile soils (I think you mean the sandy Alfisol) are suffering the most from MP? I guess, that soils with a high capability of aggregation would be more susceptible. Please deduce your hypothesis.
ACs: Many thanks for this very good point. We modified the text from: " *The underlying hypotheses are: 1) the impact of microplastic contamination on physical soil properties and hydrological processes differs with soil type, with more marked effects in the more "fragile" ones; 2) microplastic contamination, negatively affecting the structural state of the soil, increases the extent of soil erosion*" to: "*The underlying hypotheses are: i)polyester MP fiber contamination, negatively affecting the structural state of the soil, increases the extent of soil erosion; ii) the adverse impact of polyester MP fiber contamination on physical soil properties and hydrological processes differs with soil type, with more marked effects on soils that are characterised by a low aggregation ability. In these soils the presence of MP fibers could substantially increase the susceptibility to the erosion processes*"

Material and Methods
In general: The comparison of only the t = 6 month data is sufficiant, but t = 0 data would be nice.
ACs: We agree with the Referee. Surely comparison at t=0, as well as other time points, would have been beneficial , but the amount of work would have become unmanageable.

Line 68: What is „good"?
ACs: We meant a well-structured soil. We changed the text from: "*a good and stable structure*" to: "*The sampled soil is well-structured, with a clay texture…*". We have modified this part by firstly reporting the general characteristics of the three soil types and then the specific traits of the sampled soils

Line 69: What are „other elements of fertility"?

ACs: We specified the other elements of fertility: "*(P, K, N, etc)*"

Line 81: N concentrations change very fastly after sampling due to microbial processes. Is that measurement important for the underlying work?

ACs: Since Total N is mostly organic, it does not change so quickly (so that it is usually considered as a soil fertility index). The quickly changing forms are the inorganic ones (NO_3^-, NH_4^+) which are a small fraction of the Total N. Those changes are mostly due to the uptake by plants and soil microbiome, and environmental dispersion (volatilization, percolation, etc.)

Table 1: It would be helpful to add a column „texture" using KA5 (Lt3, Ls2, Slu) or SSM.

ACs: We used the USDA soil texture classification as it is one of the most used classification systems. We added more information in Table 1 ("*Clay, Silt, and Sand were classified according to USDA (Clay < 2µm, Silt 2-50 µm, and Sand 50-2000 µm);*") and in the text: "*Particle size distribution was determined using conventional methods and soil texture according to USDA (Gee & Bauder, 1986)*."

Line 96: Are the soil macroaggregates entirely disaggregated by the blender? And do you expect significant comminution of fibers during the blending?

ACs: Many thanks for this request of clarification. We cannot exclude that the blender had an effect on soil macroaggregates. However, we think that it did not affect the robustness of our results, since the same disturbance was applied to both the control and the MP treatments and we evaluated the effects of MP fibers on soil aggregation after circa 6 months of incubation regardless of the initial aggregation status.

Concerning the effect of the blender on the MP fibers size, we have already clarified this point in the reply to the Referee #1 as follows: "We used the blender in order to provide a homogenous distribution of the plastic material in the soil. It is not so easy to chop fibers, this requires special equipment, since the fibers are very malleable. Nevertheless, before setting the protocol, we have done several tests varying the mixing time and we evaluated the fiber status using a stereomicroscope. The visual image analysis of our protocol showed that changes to fibers shape, form and size were negligible. We added the following information in the new version of the manuscript: "*We chose to incorporate the fibers into the soil using a blender to provide a more homogeneous distribution of the fibers in the soil. We tested the impact of mixing time to establish a protocol which ensured a homogeneous distribution of the polyester MP fibers into the soil, and that preserved the integrity of the MP fibers (which was evaluated through visual inspection using a stereo microscope Zeiss Stemi 2000-C; Fig. 1a and b)*.""

Figure 1, a): Why do the 3 soils have 4 colors?

ACs: We wish to thank the Reviewer for having noted this inconsistency. Experiment 1 (Impact of microplastic on soil properties) belongs to a much bigger experiment where we investigated the effects of MP fibers in several soil types. Here we reported only the results for the three soils in which we have also measured the soil erosion. We changed the fig.1 leaving only the soils used in the present study

Line 108-109: I think, a „dry-out event“ applied to all samples is no problem in this case, and I agree with referee #1 to leave that out.
ACs: We have removed this part from the revised version of the manuscript

Line 112-113: Why 8 control replicates and only 4 with MP?
ACs: We are aware that we have an unbalanced design. As we analysed our data with dabestR package, the treatment units have to be drawn repeatedly to be paired up all with the control units. This increases the statistical power to detect patterns in the data. We appreciate the clear visualization and identification of the direction of treatment effects that we can obtain from this method. To evaluate the bootstrap results, we decided to run "classical" statistical models with a strict p-value cutoff; for this we used the "nlme" package (Pinheiro et al., 2018) with implemented varIdDent() function. When both approaches come to the same conclusion, we must assume that this pattern is robust

Line 114-141: This section is a bit confusing. Why do you measure so many θ values and use van Genuchten, if you only need the saturated value, field capacity and the permanent wilting point? What is „the value determined at h=-1 m after oven-drying“ (after oven-drying, there is pF 6.5)? Why is there an oven-drying in between (which probably disturbs binding factors within the soil aggregates), and can you ensure that the re-packing of samples do not destroy soil structure (and thereby alters the soil hydrological properties)?
ACs: Many thanks for this request of clarification. The van Genuchten relationship was fitted to several measured θ values to have estimates of volumetric water content at saturation, -0.1, -1 (i.e., field capacity) and -150 m (i.e. permanent wilting point) that accounted for a larger experimental information and can therefore be considered less affected by the experimental error and hence more reliable.
We changed the text from "*The volumetric water content, θ (m^3 m^{-3}), at each equilibrium stage was calculated by adding the drained volumes to the value determined at h = -1 m after oven-drying* " to "*The volumetric water content, θ (m^3 m^{-3}), at each equilibrium stage was calculated by adding the drained volumes to the final θ value corresponding to h = -1 m that was determined by oven-drying the samples at 105°C*".
We agree that an oven-drying in between the hanging water columns and the pressure plate measurements could be a factor of disturbance. However, we decided to use relatively large sample volumes (ca 100 cm^3) for the tension plate apparatus in order to have a more accurate estimation of the possible effects of soil aggregates in the wet range of the soil water retention curve (i.e., from saturation to -1 m). At lower pressure head values, the soil structure has much less influence and water retention is mainly determined by soil texture that is not affected by oven-drying at 105 °C. In any case, to verify that the re-packing of samples did not alter the soil hydrological properties,

determination of volumetric water content at *h* = -1 m was included in pressure plate experiments for comparison with the *θ* value measured at the same potential in the tension plate apparatus. Overall, the differences between the *θ* values determined at *h* =-1 m by the two applied techniques were negligible

Lines 135-136: Macroporosity and air capacity are both defined as the pore space of macropores (>50µm), which are permanently drained due to their low matrix potential. This pore diameter corresponds to pF 1.8. Please explain your choice of pF 1 (0.1 m hydrostatic head) and pF 2 (1 m hydrostatic head).

ACs: We are aware of the different suggested classifications to distinguish between macropores and meso/micropores. However, we decided to use the well-known and accepted definition given by Reynolds et al. (2002) which suggests that macropores are pores that drain at a pressure head of -0.1 m (corresponding to an equivalent diameter of 300 µm)

Line 143: Here, the measurement of soil structure needs a clarification. What is the parameter? (I think, water stable plus water labile aggregates?)

ACs: We modified the terminology and replaced "*soil structure*" (which is a vague concept) with "*soil aggregation*"

Line 144: Dry-sieving can destroy macroaggregates especially in sandy soils. The subsequent application of wet-sieving to the same samples can lead to artifacts, as the less stable WSA could have been destroyed and the %WSA reduced.

ACs: Many thanks, this is a really good point. We agree and we would have liked to analyse different soil subsamples, however the limited amount of soil per each experimental unit forced us to repeat the measurement on the same soil sub-sample. In any case, aggregates that were destroyed already during the dry-sieving are certainly not stable during the wet-sieving (which is exactly the same as the dry-sieving plus the water valve opened). Moreover, as we compared the effects of microplastic within each soil type, possible artefacts due to this methodological approach would have very limited effects. In addition, our goal was to investigate on the one hand the particle size distribution (obtained through the dry-sieving) and on the other hand the water stability of such aggregates, which we investigated through the wet sieving. Moreover, we quantified the newly formed aggregates and their stability in water by investigating the fraction >600 µm

Line 146: If the soil macroaggregates were entirely disaggregated, also the other size classes >250µm give information on newly built macroaggregates.

ACs: We agree on the importance of this information and we have decided to add the MWD (both dry and wet) in the "*Results*" section and the aggregate classes (both dry and wet) in the "*Supporting Information*"

Line 154: The WSNFA(%) is a tricky parameter. The oberserved increase of WSNFA(%), that comes along with the decrease of NFA(%), can be caused by both, the increase of WSA or a loss of water labile aggregates (WLA) faster than the loss of WSA. The different implications of these two cases should be clearly discussed. Please also reconsider your choice for the WSNFA indicator.
ACs: We have already clarified this point in a reply to a comment from the Referee #1 as follows: "*We agree with both Referees and we decided to remove the "stability indices" calculated on both new-formed aggregates (WSNFA(%)) and MWD (Sindex). We are now aware that those indices could have led to misinterpretation. We decided to report data obtained with both dry sieving and wet sieving (now Fig. 5 and 6) and added the references about the percentage of newly formed aggregates and MWD*"

Equations (1), (5) and (7): Why „A" and „P" for weight instead of m?
ACs: We changed the "A" and "P" with "W" (weight)

Equation (6): Please explain the additional information given by the S index.
ACs: Please, see response above (see response to: Line 154) about S index

Line 169: Why „Therefore, …"?
ACs: Thanks. "Therefore" was deleted

Line 200: It might be helpful to add that 33.4.mm h-1 is a heavy rain event.
ACs: Rainfall intensity of 33 mm h^{-1} is common in our environment and not particularly heavy. Therefore, we preferred to not modify the text.

Results
Additional Table: What do you think about an additional table containing the trends of all measured soil properties in both experiments?
ACs: Many thanks for this good suggestion. We added the MWD (both dry and wet) in the "*Results*" section and the aggregate classes (both dry and wet) in the "*supporting information*"

In general: Is the degree of aggregation after six months within the control samples similar to the degree of aggregation observed in the field? If not, there might be further development of soil structure. Please consider that in MP samples, due to the short term approach, a delayed development of soil structure towards uncontaminated samples cannot be excluded by the experiments. This is important for assessing the environmental risk.

ACs: We evaluated the effects of microplastics after an incubation period of about 6 months (which, to our knowledge, is already one of the longest experiments in the literature currently available). We are aware that the effects of microplastics could change over time and that it would be necessary to conduct studies even with longer incubation times (months and even years)

Discussion

In general: The relation between soil texture, soil structure and the influence of MP on aggregation and water holding capacity on the one hand and between the results of the first and second experiment on the other hand are only sparsely discussed by the authors. Starting with a clear-cut hypothesis, the observed alterations should be discussed in terms of their consistency. E.g., the loss of total aggregation in the sandy soil comes along with a loss of macroporosity/air capacity and an increase of water holding capacity, which increases the ratio of percolation to surface runoff and, thus, reduces erosion. What about the more loamy soils? What could be the effect on the landscape scale?

ACs: Many thanks for this comment. We agree with the reviewer. We tried to put all the results in a single frame that establishes a link between structural, hydrological and erosion data. We added the following paragraphs: "*The data therefore showed that contamination with polyester MP fibers modified only a little and not for all soils the soil hydrological behaviour (rainfall partition into surface runoff and percolation) but it affected the soil erosion phenomena. Even this influence was moderate or even negligible when the soil was inherently little erodible. Instead, it became appreciable in those cases in which the erosion phenomena were noticeable.*

At the end of the rainfall experiment, we observed differences between the Ctr and MP runoff plots at the soil surface. In the plots of the Ctr treatment, micro-rills oriented parallel to the slope appeared during the rainfall-runoff event, especially in the contact zone between the soil and the box walls, or small cracks developed soon after rainfall, depending on the soil type (Fig. 9). Instead, neither micro-rills nor cracks were ever detected in the runoff plots of the MP treatment. Therefore, it seems that the polyester MP fibers performed a soil particle binding action, possibly microbially-mediated, that likely induced a decrease of soil erodibility. This decrease was not suggested by the sieving experiment, probably because mechanical impact of sieving cannot be considered equivalent to the impact of the raindrops (Fox & Le Bissonnais, 1998; Loch, 1994).

The smoothness of the rills that appeared close to the walls of the box implied less resistance to flow and hence a higher flow velocity. Moreover, the soil surface after rainfall generally appeared smoother in the Ctr plots than the MP plots and, especially in the darker soil, the polyester MP fibers were noted to form a diffuse fluff on the soil surface by the end of the experiment (Fig.9). Therefore, polyester MP fibers appeared to generally induce a greater resistance of the soil to flow tractive forces. An additional possible reason for the observed results was that these fibers were exposed to some degree as a consequence of erosion during the early stage of the experiment but they remained entrapped by the *subsoil. Therefore, the soil surface of the MP plots appeared to have an additional micro-roughness compared to the Ctr plots. Consequently, flow velocity and sediment transport capacity were likely greater in the Ctr plots than the MP ones (G.-H. Zhang et al., 2011). Perhaps, the fluff formed by the exposed fibers also contributed to limit rainfall detachment, acting as a mulch*."

Line 289-290 and 292-293: The Entisol (Ls2) shows alterations very similar to the Vertisol (Lt2). Although both soils differ in their clay content, they are loams, that have a tendency to build stronger soil aggregates. In contrast, the Alfisol (Slu) has a low clay content. Sandy soils do not aggregate very well. Perhaps the explanation of the effects rather benefits from this distinction then from putting Entisol and Alfisol into one class.

ACs: Perhaps some similarities can be detected between two soil groups for some properties but not for others. We avoided to gather the two soils together due to a recognition that soil texture does not play a central role on the observed properties and processes

Line 306-307: Why should MP, that is occluded within microaggregates, reduce their ability to bond with other microaggregates?

ACs: Zhang and Liu (2018) (already cited in the manuscript) found a higher amount of MP into the microaggregates rather than into the macroaggregates; and they state: "*According to the model of soil aggregation described by Edwards and Bremner (1967) and Tisdall and Oades (1982), this result suggests that the fibers-enriched micro-aggregate could not be favorably bound into the macro-aggregate (N0.25 mm).*". This is an hypothesis which can help us to explain our results and we decided to include it in our manuscript

Line 343: Is there a possibility that microplastics prevent soil erosion without having a positive effect on the aggregate structure, e.g. by simply lying around in the way of the runoff?

ACs: Many thanks for this really good point. We improved the interpretation of our results in the revised version of the manuscript. We included those aspects in the above response (see response to "*Discussion*" in general) with the modified paragraphs

Conclusion

From my point of view, the conclusion should contain considerations about the effects on the landscape scale (because we have contaminated the whole world with microplastics), long term effects and the role of lower MP concentrations (because common concentrations are some orders of magnitude below the applied 0.5 % w/w, https://soil.copernicus.org/articles/6/649/2020/).

ACs: Many thanks for this comment. We added the following sentence: "*Although the current MP contamination level in agroecosystems is some orders of magnitude below the concentration applied in our experiment, in some areas, it is steadily increasing (Büks & Kaupenjohann, 2020); therefore it is of key importance to* *investigate such contamination levels which may represent future scenarios, as is common practice in global change biology.*".

As far as it concerns the limits of our investigation with reference to the scale issues, we report here the same response that we gave to the Referee #1: "The investigation was performed at the microplot scale and we hesitate to extrapolate the results to larger scales since active erosion processes are scale-dependent. We suggested that our results could help to delineate the hypotheses that deserve consideration in larger scale investigations. We clarified this aspect in the text. The following part was added in the revised version of the manuscript: "*We are aware that active erosion processes vary with the measurement scale (Cammeraat, 2002). At the microplot scale adopted in this investigation, erosion is expected to be due to the rainfall impact and the interrill flow (Bagarello & Ferro, 2004) and it should be a transport-limited process as a consequence of the reduced rain impacted flow and the limitation of flow velocity (Boix-Fayos et al., 2006; Chaplot & Le Bissonnais, 2000). Therefore, the collected data provide information on particle detachment and the early stage of their transport that can be expected to occur in upland agricultural soils during intense rainfall events. However, in agricultural fields, rill erosion can dominate total soil erosion due to the simultaneous occurrence of long slopes (dozens of meters or more) and the exposure of bare soil surfaces to rainfall in some periods of the year (Rejman & Brodowski, 2005). The effects of polyester MP fibers on rill erosion require specific testing but, also according to this investigation, it could be expected that rill development is hindered since the presence of the contaminant we used makes the soil intrinsically less erodible.*""