Thank you very much for your time and the thorough evaluation of our manuscript. We highly value your constructive comments and questions that we believe will help to further improve our work.

In the following, we address all comments point by point and suggest changes for a future manuscript revision (highlighted in bold face).

General

Very welcome is the point, that the authors says, that this is a „first screening“ and not a final result (including a worldwide calculation) for the rest of the world. Therefore I would avoid the deeper comparison to other studies (especially to Dierkes 2019), especially when other techniques were used. Ever through these authors did not interpret their work as a „snapshot“, the goal of this article should be this „first screening“. Furthermore the study should focus on the results and not on the comparison of the methods (Who is the best one?). Therefore, please shortens the text between L. 263-274, nobody needs this „Hunt for the lowest LOD“ any more.

> Our response: Thank you for this remark. Following your recommendation, we suggest to substantially shorten the paragraph to:

“While this clearly defines the quantitative limits of the method, our working range is still 10–100 times lower than that of previous applications involving solvent-based Py-GC/MS. Dierkes et al. (2019) and Okoffo et al. (2020), for instance, spiked 1 g of quartz sand and biosolids at 0.05–50 g kg−1 of various polymers to evaluate their accelerated solvent extraction with THF and DCM, respectively.”

Specific comments

Is there an meaningful reason for separation of particles larger than 2 mm and
subsequent analysis using ATR-FTIR or should it be better to go down with the limit value to 0.5 mm for example?

> **Our response:** Our solvent-based Py-GC/MS method was intended as a first and simple screening tool for soil-associated plastic debris that complies with the definition of fine soil (<2 mm). FTIR–ATR was used as a complement for all remaining larger particles.

The advantage of the present method (density separation, polymer extraction and detection) in comparison to the method of Dierkes (polymer extraction and detection) is the investigation of a higher field sample volume. Therefore it is expected, to get a more homogeneous, representative result. Did the authors proved this by various loading of sample volume with spiked polymers? Otherwise, please comment this more clearly and highlight this as an advantage from the beginning (not in the conclusion!)

> **Our response:** We did not assess various sample amount for sample homogeneity but simply aimed for a maximum sample size from the beginning on. To acknowledge this, we suggest to move this to the introduction instead of a results (Line 48ff):

“To better account for the heterogeneous distribution of plastic debris in soil, we further refined and validated a new sample preparation procedure involving soil aggregate dispersion and density separation that allowed for the analysis of up to 50 g soil.”

L.84: Please check, if paper bags contain PS signals. PS copolymers are often used for paper stabilisation and might be a source for the unclear PS signals. The reason in L 314 is very speculative and should be deleted, so far this is not documented for the investigated soil.

> **Our response:** Thank you for this important remark. We measured the paper bags with our solvent-based method. An extract with 250 µg/mL paper in trichlorobenzene/xylene, which exceeded the maximum concentration of our calibration curve, did not induce any PS or PE and PP signals above LOD. We agree that Lines 310–313 are rather speculative. However, such information may be a good starting point for other researchers to conduct a follow-up study on this issue. We suggest to modify the sentence in Line 312 to better communicate the underlying uncertainty of our statement:

“In the past, beads made from expanded PS were used for the conditioning and stabilization of horticultural soils (Maghchiche et al., 2010). However, it remained unresolved whether this was the case for the agricultural field investigated in this study.”

L.104-110: No data from MS of TGA/MS are presented. Therefore please delete this as an information (just TGA).

> **Our response:** We changed TGA/MS for TGA throughout the manuscript and suggest to modify Lines 104–109 as follows:

“DSC and TGA measurements were conducted in accordance with David et al. (2018). In brief, DSC was applied between −50 and 250 °C (10 K min⁻¹ ramp, 50 mL min⁻¹ N₂ flow, Q1000, TA Instruments, New Castle, US) to determine the
melting and crystallization temperatures of the agricultural plastic films. For the determination of polymer degradation onsets, plastic samples were subjected to TGA (STA 449 F3 Jupiter, Netzsch, Selb, Germany). The heating ramp was 5 K min\(^{-1}\) from 40 to 1000 °C under a 20 mL min\(^{-1}\) Ar flow. The degradation onset was determined by the temperature at which the polymer starts to thermally decompose (<1 % mass loss).”

L.160-162: No results from measurements using PET, PMMA, PVC, TWD are given. Please delete this information.

> Our response: PET, PMMA, PVC, and TWD were used to assess whether particularly high contents of those four polymers interfere with the selective quantification of our target polymers PE, PP, and PS. The results are reported in Line 234f and Table 3 (Interference). We suggest to rephrase this sentence to make the main message clearer:

“A LUFA 2.2 soil containing each 40 µg g\(^{-1}\) of potentially interfering, non-target PET, PMMA, PVC, and TWD did not induce significant false positive detections of PE, PP, or PS.”

L.184: Thermoanalysis include DSC, TGA and also others methods (DMA, Rheology etc.). In this present meaning it is related to DSC. Make this more precise, to avoid misunderstanding.

> Our response: Corrected:

“Complementary DSC analysis showed crystallization temperatures at 114–116 °C and melting temperatures at 158–160 °C. Between 381 and 400 °C, the polymers started to decompose into methylalkenes characteristic for PP (Tsuge et al., 2011, Fig. A2a for an exemplary pyrogram).”

L.185, 193: Please define what the meaning of degradation onset’s means. Include the determination of this value.

> Our response: We suggest the following explanation:

“The degradation onset was determined by the temperature at which the polymer starts to thermally decompose (<1 % mass loss).”

L.199, 212: BHT is not a common additive for polymers. It is as a antioxidant to small and tends to migrate from the polymer bulk. The observed signal is probably related to the thermal decomposition product (or because of degradation process) of Irgafos 168 or Irganox 1010, etc.

> Our response: Thank you for this important remark. We implemented your suggestion as follows:

“In addition, the PP fleeces from sites 1 and 2 as well as the PE perforated foils from sites 4–8 contained traces of a di-tert-butylphenol (for instance CAS
96-79-4) which is an indicator for antioxidants (Hahladakis et al., 2018).”

L.199: please replace slip agent by lubricant, is more common.

> Our response: Corrected.

L.201-202: The Non-identification of pesticides are very surprisingly: Are they used during the period of agriculture? Or are they expected as a additive in the plastic materials?

> Our response: To our knowledge, the investigated covers were not marketed with added pesticides. However, it is common practice to continue pesticide applications while the agricultural covers are in place. For that reason, we also wondered why we did not find traces of those pesticides on the covers. As we discuss in Line 201f this was “.. probably due to the limited sensitivity of the qualitative analysis and/or their low thermal stability.”. Yet, the screening of pesticides was not the primary goal of our study but we found this an interesting side note worth sharing with the scientific community.

L. 215pp: please comment the very intensive signals about 1000 cm-1 in the ATR-FTIR spectra, which are not related to PE or PP.

> Our response: We added the following sentences for clarification:

“The indistinct band between 1200 and 900 cm−1 may be attributed to C−O stretching in alcohols, acids, or ethers originating from a contamination with SOM or plastic aging (Fu et al., 2021).”

Used literature:


L.230pp: The low recovery rate of PS is surprisingly, because PS is the best soluble polymer compared to PE and PP. The explanation in l. 257 is very speculative and needs a approval. I expect, that part of the PS degrades to smaller PS oligomers or monomers, which does not fit to the calibration signals of reference measurements. PS is very sensitive to depolymerisation (Ceiling Temperature!), therefore I expect a significant lost of signals due to degradation during density separation and extraction processes.

> Our response: In our previous study (Steinmetz et al., 2020), we applied a similar solvent-based Py-GC/MS approach on the same reference soils but without prior density separation. We obtained PS recoveries ranging from 77 to 119 %. Given that we also dissolved our PS standards prior to Py-GC/MS analysis, we assumed the additional density separation to be the driving factor for the low PS recovery. However, we used saturated NaCl solution for density separation which we expected to have a negligible influence on PS depolymerization. From our perspective, this leaves polymer–mineral interactions as the most likely explanation which is also in line with the discussed literature.
L.275pp: Are all measurements using PY-GC/MS realised only once, or are there repetitive measurements at single samples?

> **Our response:** Since we designed our study to be a first screening, we ran single measurements only. We suggest to further clarify this in the methods section (Line 136f):

“*Each sample was measured once as described in Section 2.3.*”