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QUANTIFICATION OF MACRO-AND MICROPLASTIC POLLUTION OF FALLOW GREENHOUSE FARMLANDS: CASE STUDY IN SOUTHEASTERN HUNGARY

PhD Thesis

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1. INTRODUCTION

Plastic has become a major consumable product in agriculture because it has been used for better space management and growing crops in extreme climate conditions, such as in areas with low temperatures, high rainfall, or frequent dry periods. Indeed, the application of plastic materials in the agricultural sector is required from the nursery stage to the postharvest stage, thus, agriculture and agriculture-related sectors worldwide consumed 2.25 million tons of plastics in 2002. In Europe, 3.4% of 51.2 million tons of converted plastics were used in protective cultivation, e.g., greenhouses, mulching, packaging, nurseries, and propagation (Plastic Europe, 2020).

Polymers have become essential products in greenhouse industries in the form of sheets, water pipes, strings, and other materials. Plastics in greenhouses have a short lifespan; thus, until recently, they have been modified to last for ca. 3 years. Decades ago, plastic films were manufactured to last 1–2 years only. Therefore, extensive greenhouse farming generates plastic waste in large quantities and pollutes municipalities and farmlands. Plastic contaminants could be categorized based on their sizes, classifying macroplastics (> 5 mm in diameter) and microplastics (≤ 5 mm). In addition to size, plastic contaminants are made up of different structures, shapes, and colors. The prominent shapes found in the environment include fiber, film, fragments, beads, pellets, foam, etc. Abandoned greenhouses appear to have higher contamination than active greenhouses, and the pedosphere is the major receiver of plastic waste at both macroplastic and microplastic levels.

Despite the importance of greenhouse farming in the plastic pollution of the environment, there is a knowledge gap on the actual level of macroplastic and microplastic contamination in the greenhouse soils. Recent studies that tried to quantify the level of plastic pollution in the environment were mainly focused on the aquatic ecosystem. Few studies that were conducted on the agricultural environment were mainly conducted on mulching and conventional farmlands. Thus, there is a paucity of information on plastic contamination and fragmentation in greenhouse farmlands. Also, there is still missing knowledge on the level of microplastic pollution in the soil profiles, especially beyond 100 cm depth, and on the influence of soil physical properties on the vertical migration of plastics. Therefore, this study has the following goals: (1) Quantification of macroplastic litter in fallow greenhouse farmlands of southeastern Hungary. To achieve this goal, the following objectives were undertaken (a) quantifying the level of macroplastic litter on the surface of greenhouse fallows, (b) examining the morphological types of macroplastic litter in the area, (c) Identifying the sources and types of macroplastic litter in the areas, (d) studying the level of macroplastic litter fragmentation to microplastic. (2) Assessment of microplastic pollution in the greenhouse soil surface and profiles. This goal has been achieved by (a) quantifying the microplastic pollutants in the surface and sub-surface layers of soils of greenhouse fallows, and (b) examining the morphological structures of microplastic pollutants in the area. (3) Evaluation of vertical distribution of microplastic pollution in the soil profiles. To achieve this goal, the following objectives were considered: (a) quantifying the level of microplastic pollutants in the soil profiles, (b) determining the spatial distribution of microplastic pollutants in the soil profiles, (c) comparing the relationship between depth and microplastic availability, (d) describe the morphological structures of microplastic pollutants in the profiles, and (e) examine the influence of soil physical parameters on the distribution of microplastics.

2. MATERIALS AND METHODS

2.1 Sampling

Three abandoned greenhouse farmlands were selected to quantify the actual level of macro- and microplastic contamination. Sampling was performed on fallow greenhouse farmlands. As the structures of the plastic greenhouses were similar, the parcel sizes were also comparable (Szeged: 470 m^2; Szentes: 440 m^2; Szarvas: 500 m^2). Systematic random sampling was applied on 3-3 parcels at Szeged and Szarvas, and 2 parcels at Szentes. Two observers picked and collected all visible macroplastic debris on the surface of the entire selected parcels. The selected parcels were equally divided into two parts. In each part, the soil layer was divided into two layers (0–20 and 20–40 cm). Four samples from each layer were collected, homogenized, and comprised of a composite sample; hence, one composite sample represents half of a plot. In this way, soil samples were collected from greenhouse farmlands and control sites. Besides, a drill was dug in the middle of each selected parcel for soil profile samples. Soil samples were collected at 20, 50, or 40 cm intervals down to the level of the groundwater. The samples were wrapped with aluminum foil before being stored in bags and transferred to the laboratory.

2.1 Laboratory work

2.1.1 Preparation of macroplastic samples

The collected macroplastics were cleaned by submerging the contaminants into buckets containing tap water and soaked for 48 hours. The water used for cleaning was passed through a 5 mm sieve to collect all macroplastics. The larger and retained plastic materials were combined and dried for 4 days at room temperature. Subsequently, the macroplastics were separated, counted, and measured based on their size, shape, color, polymer composition, and possible source types (agricultural and nonagricultural). All morphological categories were counted and weighed. Approximately

10% of the macroplastic pieces were taken to a Raman spectroscopic analysis for polymer composition identification.

2.1.2 *Preparation of microplastic samples*

The soil samples were oven-dried (40°C), gently ground into smaller pieces, and sieved with a 5 mm sieve. Digestion of soil organic matter was performed using 30% H_2O_2, and Fenton reagent. The solution was heated at 70°C until all liquid evaporated. Consequently, $ZnCl_2$ [1.5 g/cm^3 (5 mol/L)] was used as a flotation salt. The complete solutions were shaken for 1 h at 200 rpm in an orbital shaker, after which they were settled for 24 h. Approximately 20 ml of the upper supernatants were collected with a glass pipette and $ZnCl_2$ was added to the solution, which was shaken for 30 min in the orbital shaker for a second time. The upper supernatants were again collected and combined with the first supernatants to form single microplastic extracts. The extracts were filtered through a nylon membrane filter (20 μm) and Whatman filter (0.45 μm) using a vacuum pump. The filters were placed in Petri dishes and covered with aluminum foil, dried at room temperature for 2 days. The dried filter papers were put under a light microscope for microplastic quantification and characterization.

2.1.3 *Preparation of Soil physical parameters.*

Soil particle size distribution was determined using a Fritsch Analysette 22 MicroTec laser particle sizer. Soil bulk density was determined using the weighing bottle method. Saturated hydraulic conductivity was determined using both the constant water head method and the falling head method because the soil samples contained a high proportion of clay.

3. RESULTS

3.1 Macroplastic abundance

Greenhouse farming is a major contributor to the macroplastic litter in the agricultural environment; however, the degree of macroplastic pollution is highly variated related to the greenhouse farming duration, farmland abandonment duration, greenhouse size, clearing activities, and climate factors. Macroplastic debris was discovered in all sampled parcels of the fallow greenhouse farmlands: the overall mean abundance of 2655 pieces/ha equivalent to 5 kg/ha of macroplastic was scattered on the surface of greenhouse farmlands. The maximum mean abundance (4328 pieces/ha) was found in Szeged (Figure 1), and the second-highest abundance (3513 pieces/ha) was found in Szarvas; whereas, Szentes had the lowest abundance (125 pieces/ha). Likewise in terms of weight, the highest contamination was found in Szarvas (10.7 kg/ha), followed by Szeged (6.4 kg/ha) and Szentes (1.2 kg/ha). The differences in greenhouse farming and farmland abandonment duration, greenhouse size, clearing activities, and climate may be contributing factors to this variance. The extended duration (27 years) of greenhouse farming in Szarvas may be the reason for the highest litter weight contamination.

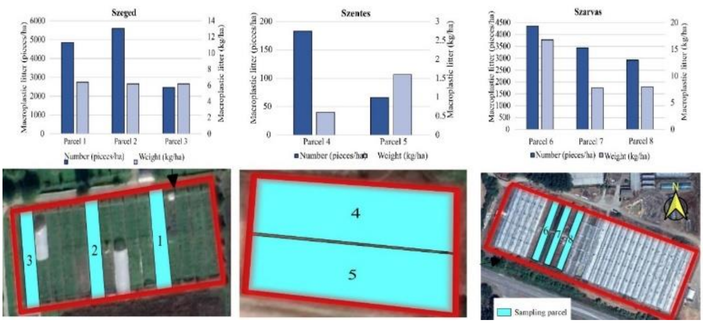

Figure 1. Total macroplastic abundance (in number and weight) at the studied parcels

3.1.1 Size and fragmentation of macroplastics

The macroplastic litter in the greenhouse environment has different sizes due to ongoing weathering, which is influenced by single or multiple factors (such as climate parameters, agrochemicals, atmospheric contaminants greenhouse structure). The fragmentation and size abundance were not uniform in the study sites. The size class of 1-5 cm was the most prevalent (46%) in Szeged, whereas >15 cm was the most prevalent (80%). The size class with the highest abundance in Szarvas was 1.0-0.5 cm (42%). The fragmentation of macroplastics into microplastics was detectable in all study areas. In Szeged, for example, macroplastic fragmentation was visible in every parcel because there were smaller macroplastic pollutants (≤10 cm) than larger plastic contaminants (>10 cm). Additionally, 300 pieces/kg of microplastic were on average discovered on the soil surface (0-20 cm). This demonstrated that the fragmentation of macroplastics is an ongoing process and that the conversion of macroplastics to microplastics had already started. Similar levels of contamination were found at Szarvas, where fragmentation was also present since smaller pieces of macroplastic were nearly twice as prevalent as larger ones. Additionally, microplastics were discovered on the surface of the local soils at a rate of 1000 pieces/kg on average. The parcels in Szentes had little fragmentation, as most of the pieces belonged to the group of larger plastics. Additionally, only an average of 125 pieces/kg of microplastics were discovered on the soil surface. Differences in weather parameters (such as annual temperature, seasonal temperature variations, solar radiation, frequency of heavy rainfall and hail), chemical usage, and thermal water heating of the greenhouses, could all contribute to an increase in the advanced fragmentation states in the areas.

3.1.2 Origin of Macroplastic Contaminants

Not all macroplastic contaminants in the abandoned greenhouse farmlands originated from the plastic materials used in the greenhouse. Unlike popular hypotheses and the previous findings that reported plastic litter as greenhouse-oriented, this study revealed that plastic litter from different sources also contributed to polluting greenhouse areas. The overall abundance showed that only 90% of the litter originated from agricultural sources, and 10% of the litter had non-agricultural origins (Figure 2).

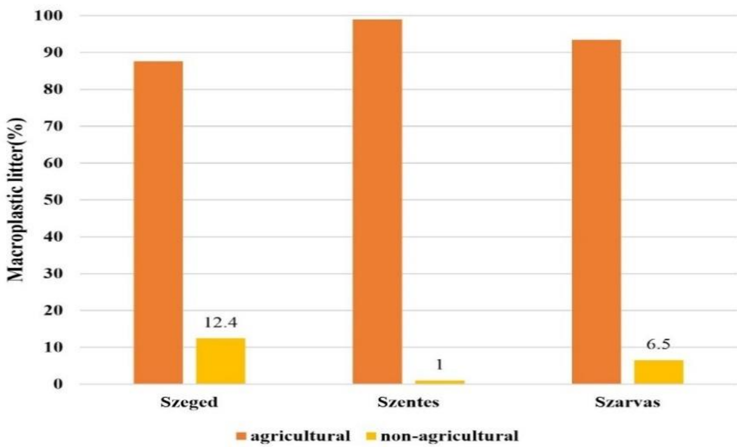

Figure 2. Abundance of macroplastics with agricultural and nonagricultural origin

Most agricultural plastics, including old films, broken pipes, and aged fibers that disintegrated as a result of weathering, agrochemicals, and other variables, were directly connected to the production of macroplastic litter. Meanwhile, litter from different sources (candy and sweet wrappers, residential litter, etc.) could be brought in from neighboring roads and rural areas by environmental elements including wind or runoff. Also, non-agricultural contaminants are present in the parcels due to unlawful trash deposition.

3.1.3 *Morphological characteristics of macroplastics*

Macroplastic litter has different structure, shape, and color (Figure 3). Film and fragments made up most of the agricultural equipment contributing 74% –95% and 15% –25% respectively. The most prevalent macroplastic color was transparent (68%), followed by black (11%). The Raman spectroscopy analysis revealed that the polymer composition of most of the plastic recovered was PE (79%–93%) and PVC (12-18%). The morphological results of the contaminants revealed that macroplastic litter occurred due to the extensive usage of plastic films in greenhouse farming as well as the weathering of plastic materials because of their low durability and surface-clogging nature.

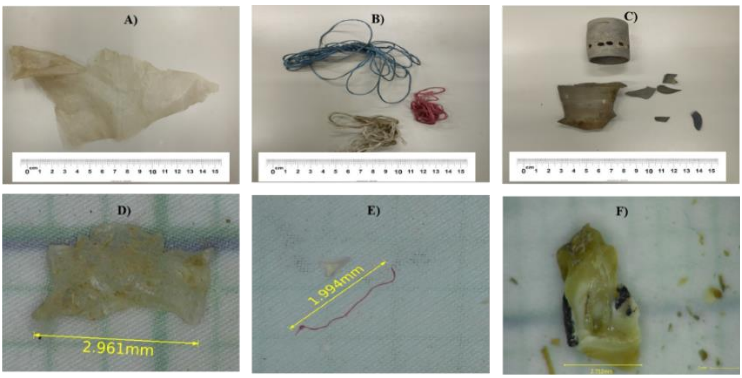

Figure 3. Some of the recovered macro-and microplastic contaminants. A-C are macroplastic (A) film, (B) fiber, (C) fragment. D-F are microplastic(D) film, (E) fiber, (F) fragment.

3.2 Microplastic abundance

Microplastic contaminants pollute not just the surface, but also the sub-surface layers of greenhouse soils, however, the surface layers were more polluted than the sub-surface. The average microplastic abundance in the entire study area was 440 pieces/kg. The average microplastic contaminants abundance was 225±61 pieces/kg in Szeged, 125±52 pieces/kg in Szentes, and 866±102 pieces/kg in Szarvas. The reason for the variation between the study areas is attributed to the different levels of macroplastic weathering and duration of abandonment. However, the distribution of the contaminants varies vertically (Figure 4). The microplastic content on the soil surface (0-20 cm) was usually higher (mean: 475 pieces /kg) than that of the sub-soil (20-40 cm: mean: 338 pieces /kg).

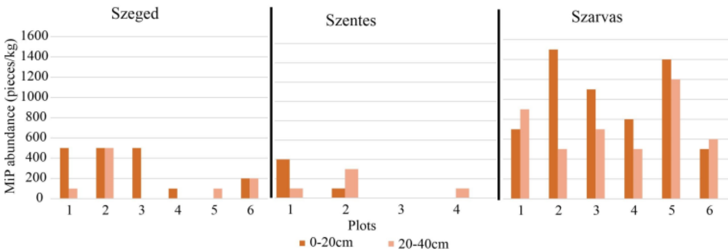

Figure 4: Abundance of microplastics in the sampling plots at 0-20 and 20-40 cm depths

***3.2.1** Morphological characteristics of microplastics in the soil surface and subsurface layers (0-40 cm)*

The distribution of microplastic types is not uniform in the soil profile: larger microplastics were recovered in the soil surface (0-20 cm) than in the sub-surface (20-40 cm) soil layer. Similarly, film and fragments dominated the surface layers while fibers were main the structure found in the subsurface. PP and PP in the form of transparent color were the major polymer compositions present in the *areas.* Considering the size of all sites, the most abundant size was 0-1 mm (49%) followed by the 1-2 mm size class (21%) in all the study areas. This disparity in the soil depths could be explained by the fact, that surface layers directly receive fragmented plastic contaminants, and only a few smaller microplastics could get into the deeper layers of the soils because of land management, soil texture, and cracks in the soil. In the greenhouse soils, microplastic fibers (44%), films (29%), and fragments (18%) are the dominant morpho-types (Figure 3). Fibers originated from the plastic fiber threads used for tightening greenhouse structures. Films and fragments originated from broken foils, pipes, crates, and other plastic materials. However, the largest film and fragment structures were predominantly found on the surface of the soils, while smaller fibrous contaminants migrated and polluted the soil sub-surface. The reason for the high abundance of fibers in the soil depths is that (1) fiber is the dominant microplastic type in the greenhouse environment, and (2) their light nature makes them easily transportable by water and they could penetrate the soil pores and cracks. Furthermore, the microplastic contaminants in the greenhouse soils are composed of different polymer compositions and a wide range of colors. The overall frequency of the polymer composition reflects that PP was the dominant polymer (44%), followed by PE (29%), PET (16%), PVC (9%), and PU (2%). Similarly, microplastics appear in a wide range of colors: transparent (35%) and blue (15%) colors were the dominant colors found in the samples. Both polymer composition and color results show the link between the plastic used in the greenhouse farmlands and microplastics. For example, PE and PP were respectively used for greenhouse coverage and tightening, thus their smaller pieces were recovered in the entire soil. Also, red fiber, black pipes, and blue plastic materials were recorded as macroplastic in the study areas and the same colors were found in the soil in the form of microplastics.

3.3 Abundance and distribution of microplastic in the entire profile

The level of microplastic distribution was not uniform vertically as some profiles contained more contaminants than others, but microplastics were not found in deeper layers than 160 cm. The average microplastic abundance varied among the analyzed profiles: 80 pieces/kg, 4 pieces/kg, and 96 pieces/kg, for Szeged, Szentes, and Szarvas respectively. However, the greatest depths microplastic appeared were 160 cm in Szeged, 50 cm in Szentes, and 140 cm in Szarvas. Predominantly, microplastics were more concentrated in the upper layers of the soil profiles than in the deeper layers. The morphological information of contaminants found in the soil depths was mostly fiber of smaller sizes with distinctive transparent colors as these materials are constantly used in the greenhouse environment.

3.4 Soil properties and vertical distribution of microplastics

3.4.1. Particle size distribution and microplastic abundance in the entire profile

The finer-grained soil layers (with a high proportion of clay and very fine silt) contained a very low abundance of microplastics; while the coarser-grained layers (with medium and coarse silt) contained a much higher amount of microplastic. The distribution of microplastic was not uniform in the soil textural classes, as medium (15.6-31 µm) and coarse silt (31-63 µm) samples contained more microplastic compared with clay (<3.9 µm) and very fine silt (3.9-7.8 µm) soil samples. Furthermore, the soils were further categorized into four textural soil classes: silt loam, silty clay loam, silty clay, and clay loam. Of which silt loam had an average of 200±48 pieces/kg microplastics, while the silty clay loam had an average of 77±26 pieces/kg. However, there was no record of microplastic distribution in silty clay and clay loam textural classes. The presence of microplastics in some parts of the studied profiles could be partly explained as a result of the coarseness of the soil, partly by the presence of cracks in the soil as it was widely observed in the areas during the fieldwork.

3.4.2. Porosity, bulk density, saturated hydraulic conductivity of the soils, and their effect on microplastic distribution

Microplastics could vertically migrate downward through the pores and cracks to the soil; however, the beneath layers could stop the movement of water and microplastic because the rate of penetration in these compacted layers is very little or not possible. Concerning the soil porosity and bulk density, the studied soils can be categorized as normal; thus, the movement of water and contaminants especially through the surface layers is possible. Most of the non-compacted layers were recorded close to the surface, while the compacted layers were recorded in the deeper soil depths (50-60 cm, 70-80 cm, 80-90 cm, and 100-110 cm). Hence, the movement of microplastic was recorded especially in the porous layers. On the other hand, the low saturated hydraulic conductivity nature of the soil made the microplastic migration difficult, except in one sample in Szeged (P1: 30-40 cm) where the soil retained very few microplastic contaminants (66 pieces/kg) and let pass plentiful microplastic contaminants (133 pieces/kg). In general, the restriction of the downward movement of microplastic was observed in the deeper layers where the level of soil compaction was high. However, since the study reveals that the soil's physical properties did not support favorable conditions for vertical migration of the microplastic contaminants because of the compacted layers in the soil depths, probably, other processes such as soil cracks support the migration of microplastic contaminants through the soil layers.

PUBLICATIONS RELATED TO THE RESEARCH

Saadu I., Farsang A., Kiss T., (2023), Quantification of macroplastic litter in fallow greenhouse farmlands: a case study in southeastern Hungary. Environmental Sciences Europe 35:63.10.1186/s12302-023-00777-6
(Journal's Impact Factor **5.481)**

Saadu, I., Farsang, A. 2023. Plastic contamination in agricultural soils: a review. Environ. Sci. Europe, 35, 63. https://doi.org/10.1186/s12302-023-00720-9
(Journal's Impact Factor **5.481)**

Saadu, I. and Farsang, A. 2022. Greenhouse farming as a source of macroplastic and microplastic contamination in agricultural soils: A case study from Southeast Hungary. Agrochemistry and Soil Science, 71/1, 43-57. 10.1556/0088.2022.00120
(Journal's Impact Factor **0.1)**

Saadu I**.**, Farsang A., (2022), Microplastics in the soil profile and groundwater of greenhouse farmlands of Southeast- Hungary. Book of Proceedings, International Scientific Conference GEOBALCANICA, Serbia. DOI: https://doi.org/10.18509/GBP22035s

Saadu I., Farsang A., (2022), An economic method of microplastic separation, extraction, and identification in agricultural soils. PROCEEDINGS OF THE 27th International Symposium on Analytical and Environmental Problems Szeged, Hungary November 22-23, 2021. Pp. 24- 28. ISBN 978-963-306-835-9

Co-authors' waivers

I, Dr. Tímea Kiss, as co-author of the publication entitled "*Quantification of Macroplastic Litter In Fallow Greenhouse Farmlands: Case Study In Southeastern Hungary*" published in the journal "Environmental Sciences Europe" officially declare that the jointly published results in the thesis and the publication are greatly contributed by the candidate and was not or will not be used in the past or in the future, respectively, for the purpose of acquiring an academic degree or title.

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