UNIVERSITY OF SZEGED

FACULTY OF SCIENCE AND INFORMATICS

DOCTORAL SCHOOL OF GEOSCIENCES

DEPARTMENT OF GEOINFORMATICS, PHYSICAL AND ENVIRONMENTAL GEOGRAPHY

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

Ph.D. Thesis

SAADU, Ibrahim

Supervisors:

Dr. Kiss Timea

Prof. Dr. Farsang Andrea†

Szeged

2024

Table of Contents

List of abbreviations

HDPE: High-density and medium-density polyethylene

PA: Polyamide

PE: Polyethylene

PET: Polyethylene terephthalate

PMMA: Polymethyl methacrylate

PP: Polypropylene

PS: Polystyrene

PU: Polyurethane

PVC: Polyvinylchloride

SEM: Scanning electron microscope.

SOM: Soil organic matter

1. INTRODUCTION

Environmental pollution by plastic contaminants is an emerging and one of the hot topics of discussion by researchers and environmentalists in the 21st century. Global plastic production has increased rapidly since the end of World War II (Pazienza and Lucia, 2020), as it increased from 1.5 million tons in 1950 to 367 million tons in 2020 (Plastic Europe 2020). After the quiescence in 2020 due to the COVID-19 pandemic, plastic production increased further to 390.7 million tons in 2021(Plastic Europe, 2022). Since their inception, dependency on plastic has made life inevitable because plastic is used in different life endeavors such as agriculture, automobiles, pharmaceuticals, construction, etc. (Hamid et al. 2018). Thus, the century is termed as 'plastic age' (Blocker et al. 2020). The most produced plastic polymers are polypropylene (PP: 19.3%), high- and low-density polyethylene (PE: 14.4%), polyvinylchloride (PVC: 12.9%), high-density and medium-density polyethylene (HDPE: 12.5%), polyethylene terephthalate (PET: 6.2%), polyurethane (PU: 5.5%) and polystyrene (PS: 5.3%). Other polymers such as fossil-based thermoplastic accounted for 7.1% (Svedin, 2020; Plastic Europe, 2023). However, the extensive usage of plastics, as well as their single-use nature led to a waste generation that ends up landfilled or discarded into the environment, because the recycling rate is very low (Yu et al. 2021). Thus, plastic contamination is recorded in all the components of ecosystems. Worldwide, the global estimation of plastic waste in the environment has already reached about 5 gigatons (Plastic Europe, 2023). Plastic contaminants are categorized into classes due to their sizes. The particle size of plastic contaminants can be macroplastics (> 5 mm in diameter), and microplastics (≤ 5 mm in diameter) (Sarker et al. 2020; Liu et al. 2018). In addition to sizes, macroplastic contaminants are made up of different structures, shapes, and colors. The prominent shapes found in the environment include fiber, film, fragments, beads, pallets, foam, etc. (Piehl et al. 2018; Meng et al. 2020).

Plastic has become a major consumable product in agriculture, because of its cheapness, impermeability to precipitation and gases, malleability, lightweight, and ability to maintain a uniform soil temperature, transport fertilizers, and control weeds, diseases, and pests (Adrady, 2003; Patel and Tandel, 2017; Sussana, 2018). Indeed, the application of plastic materials in the agricultural sector is required from the nursery stage to the postharvest stage, hence, agricultural, and agriculture-related sectors worldwide in 2002 consumed 2.25 million tons of plastics per year (Andrady, 2003). In Europe, 3.4% of 51.2 million tons of converted plastics are used in protective cultivation, e.g., greenhouses, mulching, packaging, nurseries, and propagation (Plastic Europe, 2020). The pedosphere is a major receiver of plastic waste because the production of plastic materials and landfill activities occur on the soil. Another important pathway of plastic materials to the soil is through agriculture. Agricultural soils receive primary plastic materials through the application of sewage sludge, compost, fertilizers, and agrochemicals containing compounds of microplastics and other agricultural input (Corradini et al. 2019; Katsumi et al. 2020; Meng et al. 2020).

Globally, plastic greenhouse farming covers 220,000 ha of land and consumes 250,000–350,000 tons of plastic film annually (Plastic Europe, 2019). Using plastic films is vital for better space management and growing crops in extreme climate conditions, such as in areas with low temperatures, high rainfall, or frequent dry periods (Patel and Tandel, 2017). Polymers have become essential products in greenhouse industries in the form of film sheets, water pipes, strings, and other materials. Plastics in greenhouses have a short lifespan. Decades ago, plastic films were manufactured to last 1–2 years (Giacomelli and Robert, 1993). In recent years, with the advancement of technology, the durability and performance of these plastic films have increased by up to 3 years (Dehbi et al. 2015; Babagyayou et al. 2020).

Weather variables, such as high temperature, solar radiation, precipitation, and wind were found to be among the factors responsible for the physical weathering, aging, and quality deterioration of plastic films and other plastic materials in the greenhouse system (Dilara and Briassoulis, 2000; Vox et al. 2008; Alhamdan and Helal, 2009; Babagyayou et al. 2020). Similarly, the application of agrochemicals containing sulfur, halogens, iron, and chlorine has been confirmed to cause the early aging of plastic films (Meides et al. 2022). Furthermore, environmental pollutants, such as hydrocarbon, nitrogen oxides, sulfur oxides, and particulates, are other external factors that enhance the chemical degradation of polymers by abstracting hydrogen from the polymer chain, which weakens the polymer structure and causes further depolymerization (Dilara and Briassoulis, 2000).

Extensive greenhouse farming generates macro- and microplastic waste in large quantities and pollutes farmlands in various environments (Isari et al. 2021; Saadu and Farsang, 2022). Abandoned greenhouses appear to contribute more contamination to agricultural areas than those of active greenhouses (Wang et al. 2022). In some parts of the world, unused plastic equipment is typically disposed of by burning, uncontrolled scattering in fields, or it is transported to unauthorized dumping sites (Dilara and Briassoulis, 2000; Svedin, 2020). The fragmentation of the plastic contaminants leads to the formation of meso- and microplastic waste that pollutes the agricultural environment (Svedin, 2020). Most studies on macroplastic contamination and fragmentation were conducted in aquatic environments (Weinstein et al. 2016; Kalogerakis et al. 2017; Liro et al. 2023). Only a few studies quantified the macro- and microplastics in agricultural setups, such as in greenhouses, mulching, and conventional farmlands. Most of these studies were conducted in mulch farming systems, whereas only 7% of them were conducted on greenhouse farms (Saadu and Farsang, 2023). Furthermore, the macroplastic undergoes further fragmentation and produces microplastics. The study of microplastic pollution in the environment is highly emerging because of its direct effect on the environment. Similarly, the predominant studies of microplastics have also concentrated on aquatic pollution (Ivar do Sul, 2014; Vaughan et al., 2017; Peng et al., 2018). Thus, there is a paucity of knowledge on the level and relationships of macro- and microplastics in greenhouse environments.

The environmental pollution stemming from plastic contamination has intrigued the interest of scientists and studies on the effect of plastic contamination in the soil, waters, and atmosphere. The presence of plastic contaminants affects soil quality and fertility by altering its structure, bulk density, and water-holding capacity (Liu et al. 2018; Kundu et al. 2021; Yu et al. 2021; Stefano and Pleissner, 2022). It also affects the natural condition of the soils which as a result affects the plants, surface water, and the micro- and macro-organisms of the soils. For example, Machado et al. (2018) used 2% contamination of four microplastics (polyacrylic fiber, polyamide beads, polyester fiber, and polyethylene fragments) on loamy sand soils and found that microplastics affected water holding capacity, microbial activity, aggregation functions, and bulk density of soils. In addition, microplastics adsorb and transport contaminants such as heavy metals in the soil environment (Li et al. 2019; Beriot et al. 2021). Thus, the health of soil organisms and the enzymatic activities of these organisms are disturbed by microplastic contamination (Cyvin et al. 2021). Moreover, the presence of microplastics in the soil affects the development and health condition of the soil organisms (Boots et al. 2019). Maaß et al (2017) provided a comprehensive study on how microplastic affects the mobility of soil *Collembolan* species.

Microplastics transfer vertically to the soil and soil profile through leaching and the activities of soil organisms. It can also move down through agricultural practices, such as mulching, irrigation, and greenhouse farming (Giacomelli and Roberts, 1993; Andrady, 2003; Dehbi et al. 2015; Maaß et al. 2017). The small size and fibrous contaminants have high migration potential (Zhang et al. 2018; O’Connor et al. 2019). The soil's physical structure plays a vital role in the vertical movement of microplastics from the soil surface to the soil depths. The soil properties such as infiltration, porosity, soil aggregate, soil structure, and particle size distribution not only influence the runoff but also the rate at which the soil will allow the penetration of pollutants in the soil. The clay soils that have high contents of montmorillonite minerals develop cracks easily and serve as entryways of microplastics into soil profiles (Zhao et al. 2022). Furthermore, the infiltration capacity of soils influences the migration of microplastics (O'Connor et al. 2019; Mora et al. 2020). However, the vertical penetration of microplastics in the subsurface remains unexplored because of the lack of studies. Therefore, it is necessary to study various influential parameters to better understand the complex vertical mobility of microplastics in the soil. Despite the abundance of some studies on the distribution of microplastic contaminants in the soil surface, sub-surface and profiles, there is still a paucity of knowledge on the rate of pollution in soil depths below 100 cm. Also, the information about the depth and abundance relationship is still limited.

However, the quality of agricultural products and the growth and photosynthesis of plants are altered by the presence of microplastics (Kalogerakis et al.2017; Rezaei et al. 2019; Zhao et al. 2022; Liro et al. 2023). For example, PE contaminants harm all parts of the wheat (*Triticum aestivum*) during reproductive and vegetative growth (Qi et al. 2018). Similarly, PE film residue decreases the root length and weight density in corn production, as well as affecting the water consumption capacity of the corn plant (Hu et al. 2020). Some plastics such as polylactic acid decrease the chlorophyll content in leaves and the biomass of maize (Wang et al. 2020). The addition of plastics in agricultural soils causes disturbances in the metabolic stress of plants. For example, Wu et al. (2020) used different concentrations of polystyrene microplastics on rice plants, proving that the biomass and metabolism decrease which could significantly reduce rice production. Besides, plastic contaminants were found to affect the roots of perennial plants such as juvenile lime trees (Enyoh et al. 2020).

The impact of plastic contaminants was also reported on humans and large animals. Serker et al. (2020), illustrated that plastics may cause toxicity in the food chain and cause serious health problems and death to aquatic animals. Plastic contaminants get into animal (such as ruminants and birds) bodies by direct ingestion of plastic materials or through their diets and grazing process. Similarly, smaller microplastic particles get into the plant system (especially roots and leaves) through roots and cracks (Li et al, 2020). This can be easily transferred to human and animal bodies by direct consumption of the contaminated plants. Negligible effects of microplastic on animal (earthworm Eisenia fetida) fitness were recorded (Wang et al. 2019). Microplastic contaminants were found in the feces of human and domestic animals such as poultry, sheep, and pigs (Luqman et al. 2021; Beriot et al. 2021; Yang et al. 2021; Wu et al. 2021). Further, aquatic animals such as seafood were found to consume microplastics because of the plastic pollution of the seas. Human beings consume and ingest plastic through the consumption of contaminated food, such as fish and vegetable products (Luqman et al. 2021). These contaminants carry toxic chemicals such as phthalates and bisphenol that migrate and cause problems in human cells and induce cancer, especially in children (Meng et al. 2020; Jiang et al. 2021; Mora et al. 2021; Bhuyan, 2022).

The rationale of the study

The study of plastic contamination is topical and necessary because the rate of global plastic production surpasses the rate of recovery and recycling. The motivation for this research lies in the need to know the actual nature of plastic fragmentation as well as quantify the amount of plastic pollution in the soil ecosystem. Recently, the World Health Organization (WHO) lamented the lack of studies on microplastic pollution in the ecosystem. Thus, the 2023 Environmental Day (5th June 2023) was dedicated to plastic pollution in the environment with a campaign slogan "#BeatPlasticPollution". Only a few studies in the area have quantified plastic contamination in the environment. Even the few studies that were conducted in agricultural setups were conducted on mulch and conventional farming systems. This research built on the existing knowledge and provided up-to-the-minute information about plastic contamination in greenhouse farming systems. Therefore, this research provides information for stakeholders, policymakers, farmers, and scientists on plastic contamination in agricultural lands. Moreover, it provides first-hand information on the best practices for properly dumping plastic contaminants and how these contaminants could be reused within the frame of circular economy programs. Furthermore, this research supports future monitoring studies on macroplastic and microplastic contamination in the environment.

Hypotheses

The following hypotheses related to the objectives and research questions were constructed:
The first hypothesis: The larger macroplastic litter in the greenhouse fallow lands fragment

into smaller pieces and become microplastic contaminants.

The second hypothesis: Microplastic contaminants penetrate the agricultural soils and contaminate greenhouse fallow soils and subsurface layers.

The third hypothesis: The abundance and distribution of microplastic contaminants are influenced by the depth and some soil parameters.

Aims and objectives.

The goal of this research is to quantify the macro- and microplastic pollution of fallow greenhouse farmlands in southeastern Hungary. The above goal is divided into the following objectives to better understand the spatial distribution of plastic contaminants in agricultural soils:

(1) ***Quantification of macroplastic litter in fallow greenhouse farmlands of southeastern Hungary***

The goal of this work package is to analyze the surface contamination by greenhouse macroplastic and to quantify their abundance, size, fragmentation, shape, color, polymer composition, and contaminant types.

To achieve this objective, the following research questions were answered:

(a) What is the quantity of macroplastic litter on the surface of greenhouse fallows?
(b) Does macroplastic litter fragment and contribute to microplastic pollution?
(c) What are the morphological types of macroplastic litter in the area?
(d) Does macroplastic litter have the same sources and types?

(2) Assessment of microplastic pollution in the greenhouse soil surface and profiles

The goal of this part of the research is to evaluate the level of microplastic contamination in the soils of fallow greenhouses, and to quantify its abundance, differences in the soil surface and sub-surface contamination (0-20 cm and 20-40 cm), and the morphological type.

To achieve this objective, the following research questions were answered:

(a) What is the quantity of microplastic pollutants in the surface and sub-surface layers of soils of greenhouse fallows?
(b) What are the morphological structures of microplastic pollutants in the area?

(3) Evaluation of vertical distribution of microplastic pollution in the soil profiles

The goal is to analyze the microplastic pollution in the soil profiles, to reveal the vertical differences in microplastic abundance, spatial distribution, depth, and microplastic abundance relationship, morphological type of microplastics, as well as the influence of soil physical parameters in the distribution of microplastics.

To achieve this objective, the following research questions were answered:

(a) What is the quantity of microplastic pollutants in the soil profiles?
(b) What is the spatial distribution of microplastic pollutants in the soil profiles?
(c) What is the relationship between depth and microplastic availability?
(d) What are the morphological types of microplastic pollutants in the soil profiles?
(e) How do soil physical parameters influence the distribution of microplastics?

2.LITERATURE REVIEW

Plastic contamination is a problem that affects every sub-system of the environment. Thus, the study of plastic contamination is very important and necessary, as it portrays the real magnitude of problems, and the hot spots of the problem could be highlighted. This chapter gives detailed information about the background studies in the area of plastic contamination and identifies research gaps in the scientific knowledge that need to be filled.

2.1 Macroplastic Pollution in the Environment

Environmental pollution by macroplastics is ubiquitous because plastic usage is widespread in different life endeavors such as agriculture, transportation, pharmaceuticals, construction, etc. (Hamid et al. 2018). Macroplastic accounted for 10% of the solid waste discarded on the globe. They have been widely distributed on the land, even far away from inhabited areas. The accumulation is influenced by several factors such as wind, human settlement, region, and ocean current (Barnes et al. 2009). The previous studies of macroplastics were carried out in both marine and freshwater ecosystems as huge amounts of macroplastic were recorded in the oceans, rivers, and lakes (Hidalgo-Ruz et al. 2012; Barboza et al. 2019; Kundu et al. 2021; Liro et al. 2022, 2023).

Macroplastic pollution is reported in the different components of the environment. Pietz et al. (2021) reported a total of 473 macroplastic (average of 11.8 pieces/120 m^2) in the residential, commercial, agricultural, and forested areas. The highest abundance was recorded in commercial land use. Lechthaler et al. (2020) reported the presence of macroplastics in landfills, seawater, and on the surface of rocks. In their review work, they recorded 74.4 ± 20.4 PE bottles and 7.4 ± 6.5 of other macroplastic items/m^2 in the gardens of Mexico. Meanwhile, huge concentrations of macroplastics were recovered from the soils of Norway (Cyvin et al. 2021). Similarly, 23 macroplastics items/m^2 pollution was recorded in the national parks located in the area of Mount Everest in Nepal. The result showed that the mountaineers and climbers were the main polluters (Lechthaler et al. 2020).

Most of the studies analyzing the sources of macroplastic pollution in the environment have been carried out from the contribution of natural parameters. These studies involved the influence of weather variables and the morphological characteristics of landforms and rivers. Weather variables, such as temperature, solar radiation, precipitation, and wind, were found to be among the factors responsible for the physical weathering, aging, and quality deterioration of plastic films (Dilara and Briassoulis 2000; Vox et al. 2008; Alhamdan and Al-Helal, 2009; Babagyayou et al. 2020). An increase in weather parameter rates above critical values leads to plastic bond cleavage and depolymerization. Likewise, the increase of weather parameter rates on the plastic surface leads to the generation of new compounds and the removal of some compounds that reduce the plastic strength and increase early aging (Dilara and Briassoulis 2000). Other studies attempted beyond the natural causes by identifying the macroplastic pollution sources from the contribution of anthropogenic factors. This occurs because of discarding single-used plastic materials without being recycled and through the fragmentation of larger plastic materials (Weinstein et al. 2016;

Pietz et al. 2021; Yu et al. 2021). Based on the sources of macroplastics in the environment as mentioned above, macroplastics originated from both natural and or anthropogenic sources breaking into smaller pieces and increasing the total pollution in the environment. Future studies are highly required to highlight more about the processes that lead to the fragmentation of macroplastics into smaller pieces.

Although several researchers have well reported the level of macroplastic pollution in the environment, most of the studies were carried out in marine and freshwater aquatic ecosystems (Hidalgo-Ruz et al. 2012; Barboza et al. 2019; Kundu et al. 2021; Liro et al. 2022, 2023). There is a huge knowledge gap about macroplastic pollution in the terrestrial ecosystem.

2.2 Macroplastic pollution in agricultural farmlands

Macroplastic contaminations have been reported in agricultural areas. The level of contamination depends on several factors such as the farming system and the rate of macroplastic fragmentation (Meng et al. 2020; Huang et al. 2020; Saadu and Farsang, 2022).

Macroplastic contaminations have been reported in many agricultural areas of the world. Kundu et al. (2021) reported 0.5–5.5 kg of macroplastics on a 50x30 m plot of cultivated land in the floodplain of Tanzania. Here the contaminants originated from fluvial deposition, thus, the highest weight was recorded in an area directly affected by the river. Kim et al. (2021) reported that mesoplastics occupied a very small portion of 0.3% of the total plastic contaminants in the greenhouse and mulch farmlands of Korea. Meanwhile, in mulch farmlands in China, Meng et al. (2020) reported 53.7 to 108 kg/ha of macroplastic pollution, whereas Huang et al. (2020) described an average of 83.6 kg/ha of macroplastic abundance in the horticultural fields (1x1 m) of Uygur regions of China. The pollutants originated from the fragmentation of PE mulch plastics. Liu et al. (2018) reported an abundance of 6.75 ± 1.51 items/kg on the mulch surface of the suburbs of Shanghai, China. Li et al. (2022) reported plastic pollution (6796 pieces/m^2) in mulching farmlands of China, that was ten times higher than that in the control sites. The contaminants originated from the fragmentation of plastic mulch as a result of mechanical degradation. Further, Ramos et al. (2015) reported that PE covered about 0.11 m^2 ± 0.07 m^2 (10%) of the total sample surface in peri-urban Argentina. The pollution occurred because of the weather.

Plastic pollution was also reported in European countries. For example, Piehl et al. (2020) studied macroplastic contamination in conventional agricultural farmlands in Germany and reported 206 pieces/ha of macroplastics. The area recorded had macroplastic pollution due to the contamination from the neighboring sites because of wind and water runoff. Meanwhile, Stefano and Pleissner (2022) reported much higher contamination in Germany, as they found 9247 pieces/ha of macroplastics on arable lands treated with compost that contained some macroplastic contaminants. The reason for the disparity in the contents of macroplastic as reported by the above-mentioned studies is because of the differences in the level of plastic usage, time, and climate which affect the fragmentation of larger plastic materials.

Moreover, one can conclude that the pathways of macroplastic contamination in agricultural soils come from the weathering of larger plastic materials, and climatic influence (such as runoff and wind) (Ramos et al. 2015; Astner et al. 2019; Kundu et al. 2021). Other studies reported that macroplastics get into the agricultural environment through farming systems as well as the influence of human activities (Piehl et al, 2018; Meng et al. 2020; Saadu et al. 2023). Comparatively, macroplastic pollution due to the fragmentation of plastic cover films mulching, and greenhouse is higher than the other sources (such as anthropogenic). This is because, in the fragmentation of plastic, both mechanical and chemical degradation occurs on the larger body of plastic films (He et al. 2018). However, the level of macroplastic contamination in the agricultural soil does not only depend on the plastic weathering or mechanical degradation but duration and the level of plastic usage in the area (Huang et al. 2020; Meng et al. 2020; Li et al. 2022). The duration of plastic usage contributes more to macroplastic contamination than the level of plastic usage. Higher contamination was recorded in the agricultural farmlands with a short continuous history (6-8 years) compared to a long intermittent history (30 years) (Meng et al. 2020).

Furthermore, pollution of the agricultural environment by macroplastics is not only restricted to the soil surface but it is sometimes extended to the sub-surface. Liu et al. (2018) recorded a mean of 3.25± 1.04 items/kg in the 3-6 cm deep suburbs of China. Similarly, Li et al. (2022), found 6796 pieces/m^2 of macroplastics in the 0-20 cm depth on the experimental sites of Shenyang Agricultural University, China. The abundance of macroplastics was higher on the surface compared to the subsurface. The reasons for these variations include farming systems (such as plowing), duration of plastic usage, and difference in the runoff capacity between the surface and subsurface (Liu et al. 2018; Li et al. 2022). Macroplastics transferred vertically to the soil subsurface due to the entanglement of plastics with plant roots, burrowing by rodents, and anthropogenic factors (Li et al. 2022).

While additional studies have been carried out in the field of plastic contamination in agricultural setups such as mulching, and conventional farmlands, only a few have considered the contribution of greenhouse farming in the production of macroplastics. Thus, there is a paucity of knowledge on the level of macroplastic contamination as well as the nature and rate of plastic fragmentation in greenhouse environments.

2.3 Comparison of spatial studies on plastic pollution

Several studies have reported the pollution of agricultural soil by microplastics worldwide as discussed in sub-chapter 2.2. Microplastics were not only reported in agricultural soil with a history of plastic applications (such as mulch, greenhouse, and tunnel), sewage sludge application, or compost manuring but they were also reported in agricultural areas with no history of plastic application (Piehl et al. 2018; Li et al. 2022).

This study looked at plastic pollution at the continental and national scales. The distribution of microplastic contamination studies across various continents is shown in Fig.

1. From 2018 to 2022, about 60% of microplastic contamination studies were conducted in Asia, and most studies were conducted in China (Liu et al. 2018; Li et al. 2019; Chen et al. 2020; Yang et al. 2021), followed by Japan (Katsumi et al. 2020, 2021; Grause et al. 2022) and other countries. Furthermore, Europe accounted for 29% of the global microplastic contamination studies in the said period. The countries with the greatest number of studies include Germany, Spain, and the Netherlands (Qi et al. 2018; Beroit et al. 2021; Harms et al. 2021; Tagg et al. 2022). Additionally, Africa accounted for 4% of MP studies, with one study performed in Tunisia, Tanzania, and Mauritius (Kundu et al. 2021; Ragoobur et al. 2021; Boughattas et al. 2021). Similarly, North America accounted for 4% of the studies, with studies performed in Canada, the USA, and Mexico (Hernández-Gutiérrez et al. 2022, Crossman et al. 2020). However, Latin America accounted for only 3% of the studies, with studies conducted in Argentina and Chile (Ramos et al. 2015; Corradini et al 2021). One of the reasons for the differences in the frequency of studies is because of the difference in the rate of plastic applications in agricultural systems. For example, China is the highest consumer of plastic and the country with the highest application of plastic in agriculture (Plastic Europe, 2023), thus here the highest frequency studies of microplastic pollution in the agricultural environment were recorded (Saadu and Farsang, 2023).

Previous studies dealt with microplastic pollution in different parts of the world including European countries, but no research has approached microplastic contamination in the Hungarian agricultural farmlands although the application of plastic in agricultural farmland such as greenhouse and mulch is paramount (KSH, 2023).

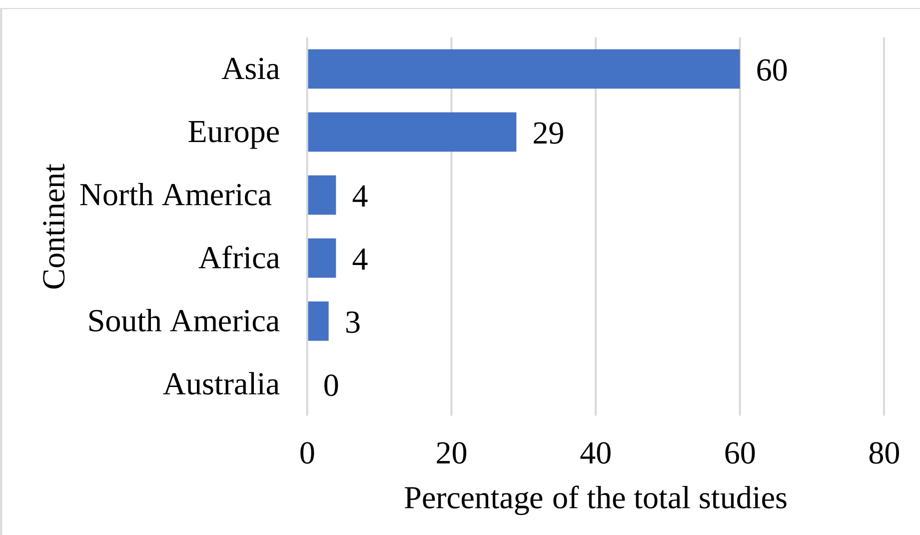

Figure 1. Worldwide distribution of plastics contamination studies conducted on agricultural land (n = 120).

2.4 Microplastic Pollution in Agricultural Soils

There is much evidence of the presence of microplastics in agricultural soils (Piehl et al. 2018; Li et al. 2019; Huang et al. 2020; Meng et al. 2020). Piehl et al. (2018) first quantified microplastic pollution in the agricultural soils in untreated conventional farmlands in Germany. After that, several studies followed it aiming to quantify the sources of microplastic contamination.

In Asia, Liu et al. (2018) studied the vegetable farmlands of suburbs of Shanghai China and detected 78.00 ± 12.91 items/ kg of microplastics in the shallow soil layers. Meanwhile, higher contamination was reported in the work of Li et al. (2019) where a total of 420-1290 items/kg was reported on soils amended with sewage sludge in the Nanjing region of China. Similarly, Yang et al. (2021) reported an average of 640 ±1250 items/ kg in the agricultural soils treated with pig manures in China. Their studies estimated the annual deposition of 3.50 ± 1.71 million items/ha. Meanwhile, Katsumi et al. (2020) reported concentrations in the range of 6-369 mg/kg (mean 144 mg/kg) of microcapsules in the agricultural soils treated with coated fertilizers in the paddy fields of Japan. Choi et al (2020) reported a mean average of 664 items/ kg in the agricultural soils (upland, greenhouse, orchard, and paddy field sites) of Yeoju City in the Republic of Korea.

The problem of plastic pollution is reported in African agricultural soils as well. Kundu et al. (2021) found 0.2-1.5 items/g in the irrigated soils of Tanzania. Much higher contamination ranges from 13.21 ± 0.89 to 852.24 ± 124.2 items/kg were recorded in the organic farming, greenhouse, mulching, and irrigated with wastewater of Tunisia (Boughattas et al. 2021). Similarly, Ragoobur et al. (2021) found an average abundance of 320.0 ± 112.2 items/kg in the irrigated soils of Mauritius.

In the agricultural soils of the Americas, microplastic pollution was also reported. For example, Hernández-Gutiérrez et al. (2022) found differences in the contents of the agricultural soils of Mexico. They found that the microplastic pollution in the mulch farmlands (400–2000 items/ kg) was greater than in non-mulch farmlands (200–1000 items/ kg) and greater than in unmanaged farming systems (200–400 items/ kg). Also, Corradini et al. (2021) found 306 ± 360 and 184 ± 266 items/ kg as respective microplastic pollution in the croplands and pastures of Chile. Moreover, Crossman et al. (2020) found an average of 541 items/ kg in Canadian agricultural soils with 5 years of biosolid application.

Some studies reported microplastic pollution in the agricultural soils of European regions as well. Berg et al. (2020) proved that microplastics accumulate in agricultural soils as a result of sewage sludge application. Thus, these soils had an average of 256% higher microplastic contents than soils without application of sewage sludge in Spain. Higher contents of microplastics (2116 ± 1024 items/ kg) were detected by Beriot et al. (2021) in the soil treated with sheep feces in vegetable farmlands of southeast Spain. Furthermore, Isari et al (2021) reported 301 ± 140 items/ kg of microplastics in vegetable farms in Greece. Schothorst et al. (2021) assessed the agricultural soils of the Netherlands and concluded that compost applications contaminate soils (2800-616 items/ kg) more than gardens and greenhouse farmlands (1253 - 561 items/ kg). Harms et al (2021) recorded 3.7 ± 11.9 items/

kg DW in the agricultural farmlands of Northern Germany. Similarly, Braun et al. (2020) detected microplastic contamination ranging from 12 ± 8 to 46 ± 8 items/ kg in the horticultural soils of Germany.

Much of the literature that considered the pollution of microplastics in agricultural soils categorized the microplastic sources into three groups: fertilization and irrigation, plastic coverage, and external sources. Fertilization occurs through the application of materials and substances to improve the quality and fertility of the soil. These include the application and amendment of soil by sewage sludge, compost, and coated fertilizer, as well as the application of irrigation water (Li et al. 2019; Katsumi et al. 2020; Yang et al.2021; Schothorst et al. 2021; Boughattas et al. 2021). Moreover, the application of plastic coverage involves the use of plastic materials for covering the plants and soils to check extreme weather conditions and conserve water and nutrients (Patel and Tandel, 2017). These involve mulch, greenhouse, tunnel, and piping (Meng et al. 2020; Yu et al. 2021; Wang et al. 2022). External sources usually involve deposition as a result of wind, water runoff, and atmospheric deposition (Rezaei et al. 2019; Allen et al. 2019; Choi et al. 2020). However, comparatively, the level of contamination differs from studies. For example, critical analysis shows that studies dealt with the fragmentation of larger macroplastics (mulch, greenhouse, and tunnel) and fertilization (amendment by sewage sludge) resulted in higher contamination of microplastics compared to other sources (Li et al. 2022; Hernández-Gutiérrez et al. 2022). These discrepancies occur as a result of differences in the sources of microplastic contamination, duration of microplastic application as well as the level of macroplastic fragmentations (Saadu and Farsang 2022; Liro et al. 2023). Other factors responsible for the differences are rainfall intensity and slope which determine the surface runoff and mobilization of microplastic particles (Zhang et al. 2022).

Even though several studies analyzed the microplastic pollution in soil mulch, soils amended with sewage sludge, compost, and coated fertilizers, irrigated soils, and soil affected by runoff and atmospheric deposition, the literature is largely underpinned on the microplastic pollution in the greenhouse farmlands despite the contribution of greenhouse farming in the production of both macroplastic and microplastic.

2.5 The abundance of microplastics in soil profiles

Table 1 shows studies about microplastic pollution in the soil profiles. Out of the ten studies reviewed, the result shows that microplastic contaminants come from different sources of which plastic mulch and greenhouse farming dominated the sources with 30% respectively. The second major contributor of microplastic contaminants in the soil profiles is sewage sludge which accounted for 20%. The other sources include animal manure and experimental sources which accounted for 10% respectively. However, the studies were conducted in different countries and China accounted for 70% of the total studies while Mauritius, Germany, and Thailand accounted for 10% of the studies respectively.

Further, the table reveals that the study of microplastic contaminants was carried out in different soil depths. Most of these studies (40%) were carried out in 20-30 cm depth. The second most studied depths were 0-10 cm and 10- 20 cm which accounted for 20% respectively. The least studied depths were 30- 40 cm and 40-50 cm which accounted for only 10% respectively. Microplastics are not only concentrated on the soil surface even though most of the studies (70%) show that the contaminants were concentrated on the surface, 30% of the studies reveal that contamination in the lower layers was higher than the surface layers. Moreover, the reason for this disparity in the soil layers occurred as a result of differences in soil management. Studies found that the level of microplastic pollution largely depends on land management (Zhang et al. 2018), for example, Schothorst et al. (2020) studied the distribution of microplastics in compost-treated and greenhouse farmlands, and their result showed that compost was found to contaminate farmland more than greenhouse farming. Similarly, Meng et al. (2020) reported the substantial contamination of microplastics in agricultural soils with high-intensity machine tillage than in soils with low intensity of tillage. Furthermore, the disparity of microplastic contamination depends on the rate of application of contaminated water for irrigation. Waters from wastewater treatment plants have been reported to contain microplastic, application of these waters may increase the contaminants load in the soil surface and sub-surface due to infiltration and percolation (Zhang et al 2018). Importantly, the presence of cracks in the soil leads to soil subsurface contamination by microplastics. This is because water, wind, and other external factors easily carry the pollutants via the cracks (Zhao et al. 2022).

However, as revealed by Table 1, the existing literature mostly covers microplastic pollution in shallow soils (sub-soil and soil profiles), but hardly exceeding 100cm depth. Therefore, there is still unexplored information about the level of microplastic pollution in the soil profiles below 100 cm depth. More especially microplastics have been recently reported in the groundwater (Su et al. 2021; Tagg et al. 2022)

Table 1: Microplastic abundance in the soil and soil profiles

Reference	Country	Plastic source	Depth (cm)	Abundance (items/ kg)		Diff.	Agric. activities
				Upper	Lower		
Liu et al. (2020)	China	Mulch	3-6	78.00 ± 12.91	62.50 ±12.97	15.50	mulch
Huang et al. (2020)	China	Mulch	40	61.9 ± 20.6	68.0 ± 41.4	-6.1	Ploughing
Zhao et al. (2020)	China	Experimental	50	7.03 g/kg	0.29 g/kg	6.74	Experimental
Yang et al. (2020)	China	Pig manure	6	84.8 ± 13.2	66.0 ± 13.9	18.8	Ploughing
Meng et al. 2020	China	Mulch	30	900	2200	-1300	Tillage
Ragoobur et al. (2021)	Mauritius	Sewage sludge/ wastewater	10-20	320.0 ± 112.2	420.0 ± 244.0	100	Ploughing/ harrowing
Yu et al. (2021)	China	Greenhouse	5–25	1443 ± 977	1312 ± 857	131	Greenhouse
Harms et al.(2021)	Germany	Sewage sludge	30	5.8 ± 8	1.9 ± 1.6	3.9	Ploughing
Fakour et al. (2021)	Thailand	Greenhouse/ Open space	20	53.2	34.6	18.6	Mulch
Wang, et al. (2022)	China	Greenhouse	10-20	826.67 ± 261.02	1073.33 ± 306.16	-246.3	Irrigation

2.6 Morphological characteristics of macro-and microplastic pollutants

Plastic pollutants are well known for their special characteristics. These include polymer composition, shape, color, and size. Different studies have reported the presence of different polymers in agricultural pollutants. Liu et al (2018) analyzed the characteristics of microplastics and mesoplastics contaminants in the agricultural soils of suburbs of Shanghai China and recorded nine polymer types. These are PP, PE, Polyamide (PA), PET, PVC, Polycarbonate (PC), Acrylonitrile butadiene styrene, Polymethyl methacrylate (PMMA), and PS. A similar result was found by Ding et al. (2020) in the Shaanxi region of western China. However, Boughattas et al. (2021) reported that there were only two dominant microplastic types in the Tunisian agricultural soils, these were: PE and Polybutyrate adipate terephthalate Schothorst et al. (2021) used Fourier Transform infrared spectroscopy and assessed the microplastic contaminants found in the soil treated with compost in Netherlands and Spain. They found that PE and PP were the main polymer compositions. Further, Isari et al. (2021) studied the polymer type of the contaminants found in the agricultural areas used for cultivating watermelons and tomatoes in Greece, their result shows that PE was the main polymer composition. To sum up, different polymer types are found in agricultural soils, PE and PVC and PP were the most common polymer types (Huang et al. 2020; Meng et al. 2020; Isari et al. 2021). The availability of polymer composition largely depends on the type of plastic used in the area and the rate of plastic fragmentation (Andrady, 2005, Wang et al. 2022).

However, several studies reported that plastic contaminants displayed in various shapes such as films, fibers, pallets, fragments, beads, and foam (Meng et al. 2020; Yu et al. 2021, Kim et al, 2021). Films and fragments were the main contaminant structures found in agricultural soils (Piehl et al. 2018; Lu et al. 2021; Mora et al. 2021). Similarly, Kumar and Sheela (2021) reported that film and fibers were the highest-recorded contaminants in the mulch soils of India. The plastic films, fragments, foams, and pellets mostly occur from the secondary sources of plastic pollution, ie. fragmentation of larger plastics as a result of the climatic, agrochemical, and pollutant influences as well as the mechanical effect on larger macroplastics (Saadu et al, 2023; Liro et al. 2023). These structures were more recorded on the soil surface compared to the soil sub-surface. This might be due to their nature of irregular shapes and structures (Liu et al. 2018, Zhang et al. 2018; Kumar and Sheela 2021). However, other shapes such as fiber and beads also occur in agricultural soils treated with sewage sludge, compost organic fertilizers, and other fertilization sources (Chen et al. 2020). In some studies, only fibrous materials were recorded in the agricultural soils (Cao et al. 2021; Kumar and Sheela 2021; Fakour et al. 2021). Other studies reported that the pollution of agricultural soils occurs with more than one contaminant structure. Fiber and fragments were the most abundant structures found in the various land-use types of China (Li et al. 2019, Yang et al. 2021; Zhang et al. 2022). Similarly, Chen et al. (2020) reported that fiber and microbeads were the major contaminants in the agricultural soils of Wuhan China. Zhang et al. (2022) found film and film as major contaminant structures in agricultural soils. Fibers mostly occur in agricultural soils from secondary sources. Recent studies have shown that fibrous contaminants have a high tendency to mobility to the soil sub-surface and profiles (Cao et al. 2021; Ragoobur et al. 2021).

Other important characteristics of plastic contaminants are size and color. These are essential in the study of plastic pollution because they refer to the sources of the contaminants and the rate of fragmentation, but only a few studies were concerned about these properties. Plastic contaminants are usually classified into different groups by size. For macroplastics, Liu et al. (2018) reported that 6.7% of the total macroplastic pollutant recovered in the mulch farmlands of China is made up of mesoplastic (5.1- 16 mm). Similarly, Meng et al. (2020) reported that 10- 50 cm^2 is the dominant size (46%) of microplastics found in agricultural soil under intensive mulching. However, Saadu et al. (2023) concluded that in the greenhouse soils, 1-5 cm size macro- film and fragments accounted for over 55% of the total contaminants. The trend is also similar in the studies of microplastics. In the studies of Li et al. (2019) and Chen et al. (2020), different categories of contaminant sizes were found in agricultural soils, but the dominant contaminant size ranges from 0- 0.25 mm. Other studies (Zhang et al. 2018; Lu et al. 2021) reported that the sizes of the contaminants reported in the mulch lands are bigger than the above-mentioned studies. Their studies found the dominant size ranged from 0.25- 0.5mm. Ragoobur et al. (2021) stated that the dominant structure of microplastic falls between 0.5- 5mm in agricultural soils treated with wastewater and sludge. The bigger size contaminants usually stay and pollute the surface of the soil (Kumar and Sheela 2021; Chen et al. 2020). While the smaller pieces migrate through the soil profile (Cao et al. 2012; Ragoobur et al. 2021, Yang et al. 2021).

Furthermore, different colors were determined in the plastic contamination. Liu et al. (2018) found only two colors in the mulch contaminants: black and transparent. Boughattas et al. (2021) assessed the color of the plastic contaminants recovered from organic farming, mulching, greenhouse, and irrigation farming and reported five different colors: black, blue, red, yellow, and green. Saadu et al. (2013) found the transparent color to be the dominant (68%) color in abandoned greenhouses. The other colors found include black, green, blue, white, and red. However, the manure compost seems to have more varieties of colors compared to other sources. For example, Yang et al. (2021) reported that compost manure contained more colors because it comprises black, blue, transparent, red, green, white, and purple with black dominating and accounting for 33%. The distribution of color systems in plastic contaminants is related to the different land uses and contamination sources. One of the reasons for the availability of different colors in plastic contamination is the contribution of external contaminations as a result of runoff and atmospheric deposition (Kundu et al. 2021; Saadu et al. 2023).

The existing studies about the morphological properties of plastic contaminants concern polymer compositions, shapes, colors, and sizes, but very few studies are concerned about the rate of fragmentation of the macroplastic contaminants to microplastics in the greenhouse environment. Also, there is a paucity of knowledge about identifying the sources of plastic contaminants using morphological properties such as color and shape.

2.7 The relationship between the physical characteristics of soils and the vertical migration of microplastics

The physical characteristics of soils play a vital role in the vertical migration of pollutants including microplastics. The transportation mechanism of soil pollutants depends on the physical properties of soil, mainly on its texture and water movement (Filep, 1998). O'Connor et al. (2019) presented that the vertical penetration of microplastics depends on several factors such as the soil particle grain size and the water cycle in the soil. The soil texture determines the soil particle arrangement and microplastic abundance in the soil. Watteau et al. (2018) observed the presence of microplastics in individualized particles in the coarsest fractions as well as some of the fine soil fractions, but they were slightly associated with the soil matrix. Yu et al. (2021) compared the distribution of microplastic in loam, sandy loam, silty loam, loam soil, and clay. Their study reported that the abundance is not uniform as soils with sandy loam contained more microplastic than other textural classes. Other studies also reported a strong correlation between soil textural class and microplastic abundance (Lu et al. 2021; Cao et al 2021).

The distribution of microplastic in soils occurred as a result of multiple effects of soil texture, bulk density, and particle size distribution (Jiang et al. 2017; Machado et al. 2018; Oßmann, 2021; Haixin et al. 2022). Notably, soil hydraulic conductivity and other moisture-soil-related properties affect the distribution of microplastic contaminants (O'Connor et al., 2019; Schell et al., 2022). The suspended microplastic particles easily penetrate the soil profiles in case of repeated wet-dry cycles (O'Connor et al., 2019; Oßmann, 2021; Schell et al., 2022). Soil with high water holding capacity and penetration ability easily allows the movement of microplastics.

However, some studies such as those made by Scheurer and Bigalke (2018) presented that soil texture has little or no effect on microplastic distribution in the soil. The reason for these differences is possible because of the differences in the agricultural practices, level, and duration of plastic usage in the areas, as well as soil amendment factors. Moreover, soil particle size distribution and their arrangement affect soil porosity and infiltration which affect the vertical distribution of microplastic (Huang and Xu, 2010; Oßmann, 2021). Concerning particle size, soil pores can be divided into inactive (< 0.002 mm), capillary (0.002- 0.02 mm), and aeration pores (> 0.02 mm). The high percentage of sand particles in the soil leads to the presence of aeration pores that cause plastic contamination in the soil (Yu et al. 2021).

Microplastic size is also an important factor that determines the rate and level of plastic migration and abundance in the soil. Compared to macroplastic and larger microplastic contaminants, studies show that smaller microplastics ($\leq$ 1mm) have the highest migration capacity to migrate and pollute the soil subsurface (Yu et al. 2021; O'Connor et al. 2019; and Zhang et al. 2022). The reason for that is, that the smaller microplastic easily moves in the smaller and larger pores and contaminates the lower soil layers. Also, fibrous contaminants were found to contaminate the soil environment more, probably because they easily move within the soil pores because of their shape and lightweight (Kumar and Sheela 2021; Fakour et al. 2021).

In addition to the soil's physical characteristics, microplastic migrates to soil profiles through the leaching of contaminated groundwater (Zhang et al., 2022). The movement of soil organisms, such as earthworms and rodents, aids in the distribution of microplastics in the environment. For example, *Folsomia candida* and *Proisotoma minuta* could transport and distribute larger particles of microplastics to a certain distance (Maaß et al., 2017). However, the cracks in the soil also serve as good pathways for microplastic distribution from the soil surface to the soil profile (Zhao et al, 2022). Importantly, agricultural activities such as harrowing, plowing, and ridging increase the high chance of vertical penetration of microplastics. Studies showed the high contamination of microplastics in areas with intense agricultural activities compared to little or no such kind of activities (Meng et al. 2020; Harm et al. 2021).

Despite some notable attempts to correlate the soil's physical properties and distribution of microplastic contaminants in the soil, there is a huge knowledge gap in portraying the actual level of this relationship.

2.8 Comparing methods of macro- and microplastic studies in agricultural soils.

2.8.1 Field sampling of macroplastics

Almost all sampling techniques applied for environmental contamination studies can be applied to plastic contamination studies. However, the applied sampling technique depends on the type of research, nature of the contamination, geomorphology, research objectives of the study, and potential contaminant sources (Thomas et al. 2020).

Different sampling strategies were employed in the studies of macroplastic pollution. Kundu et al. (2021) adopted a 50 x 30 m quadrat on a transect to collect macroplastics on the surface of agricultural farmlands. Similarly, Piehl et al. (2018) employed transect sampling by using a quadrat of 32 x 32 cm at 80 cm in between and collected macroplastic particles on the surface of farmlands. However, Ramos et al. (2015) adopted a 1m^2 quadrat and collected polyethylene contaminants in 12 sample points of horticultural soils. Meng et al. (2020) collected the macroplastic contaminants employing a 100 x 50 cm quadrat. Li et al. (2022) also collected macroplastic litter by quadrat survey method, by randomly selecting 0.5 × 0.5 m quadrats, where the soil was dug out in the block at 20 cm to collect contaminants from the subsurface. Likewise, Liu et al. (2018) collected macroplastic data by using an area of 0.5 × 0.5 m on the farmlands in a suburb. Huang et al. (2019) used random sampling by using 1 x 1 m sampling plots and collected the macroplastics in the 19 provinces of China. These studies revealed that the most commonly applied sampling technique in macroplastic studies is collecting plastic items from quadrats or transects (Liu et al. 2018; Meng et al. 2020; Thomas et al. 2020; Kundu et al. 2021). Meanwhile, most if not all studies revealed that the macroplastics were collected through hand picking of visible contaminants by careful observers who walked around the sampling sites (Piehl et al. 2018; Huang et al. 2019).

In this study, I used quadrat sampling on the rectangular parcels of used greenhouse farmlands, where I systematically selected the sampling sites. The rectangular parcels were selected because the structures of the plastic greenhouses were similar, and the parcel sizes were also comparable. Similarly, I used this method of sampling because it was used by previous studies and the results yielded positive outcomes.

2.8.2 Field sampling of microplastics

Unlike macroplastic sampling, numerous sampling techniques were used for microplastic studies. Du et al. (2020) used random sampling techniques and collected 5 samples at a depth of 10 cm within a plot of 100 m^2. Beriot et al. (2021) and Yang et al. (2021) also adopted a random sampling technique to collect the soil samples at depths of 10 cm and 20 cm. Similarly, Zhang et al. (2019), Schothorst et al. (2021) and Kumar and Sheela (2021) used random sampling techniques and collected soil at a depth of 30 cm for the analysis of microplastic contamination. However, the literature revealed that the quadrat sampling method was also adopted by many researchers for microplastic studies. Li et al. (2020) collected samples from a 0.1 × 0.1 m quadrat at a depth of 20 cm. Also, Meng et al. (2020) used the same sampling method and collected microplastic at a depth of 30 cm. Another important sampling method in microplastics is transect sampling. Wang et al. (2022)

used a transect and quadrat of 0.5 x 0.5 m to collect surface (0-10 cm) and deep (10-20 cm) soil for microplastic analysis. Kundu et al. (2021) adopted the same method and collected the soil samples from a 30 x 30 cm sampling plot. Piehl et al. (2018) adopted a 32 x 32 cm quadrat on a transect at the interval of 80 cm between them and collected microplastic data in the conventional farmland of Germany. Crossman et al. (2020) adopted the transect sampling strategy and collected soil samples at different depths (0, 5, 10, and 15 cm) from the agricultural farmlands amended with biosolid.

Furthermore, in areas where the level of microplastic contamination is not uniform and divisional difference is visible, a stratified sampling method is employed. Zhang et al. (2021) adopted a stratified sampling method and collected the soil sample on 0.5 x 0.5 m large sampling sites at a depth of 30 cm. Cao et al. (2021) employed the same method and collected a total of 85 samples at a depth of 80 cm. However, the grid sampling technique was also used in the study of microplastic contaminations. Katsumi et al. (2020) adopted the systematic grid (2 x 3m) sampling technique and collected soil samples at a depth of 15 cm. Ragoobur et al. (2021) collected a total of 40 samples at a depth of 20 cm by using 20 x 20 m grids. A combination of two or more sampling techniques may be also used depending on the nature and spatial distribution of the contaminants. For example, Wang et al. (2022) adopted both stratified and transect sampling and collected data for microplastic analysis. Archival soil data collected and efficiently stored in the past are also used in microplastic studies. For instance, Corradini et al. (2019) used soils sampled 4 years before their study and quantified the level of microplastics in regional-scale soils under different land use conditions.

The reviewed literature shows that almost every sampling type is equally adopted in the study of microplastics, but random, transect, and quadrat sampling methods are the common ones (Piehl et al. 2018; Du et al. 2020; Meng et al. 2020). The reason for their adoption may be due to their simplicity and representativeness. Also, the reviewed studies showed that for the analysis of microplastics, soil samples could be collected from different soil depths such as 0–5 cm (Piehl et al. 2018; Chen et al. 2020), 0–10 cm (Rezaei et al. 2019; Du et al. 2020), 0–15 cm (Katsumi et al. 2020; Crossman et al. 2020), 0–20 cm (Boughattas et al. 2019) 0–30 cm (Zhang et al. 2019) and 80 cm (Cao et al. 2021). The soil depth used depends on the nature of the plastic materials used, soil management techniques (e.g., plowing and harrowing), and soil physical and chemical properties (e.g., leaching, hydraulic conductivity, and infiltration capacity).

To ensure the representation and avoid sampling bias a composite sampling is applied by mixing samples from different sampling points to make one single composite sample (Huang et al. 2019; Du et al. 2020; Ragoobur et al. 2021). Meanwhile, from 4 up to 10 composite samples are combined to form 2-5 kg from which a small portion is selected for further analysis (Zhang et al. 2018; Yu et al., 2021). However, usually, a large amount of soil is collected to provide sufficient sample numbers for treatments and replication. The weight of soil samples collected for point sampling usually ranged between 0.2–3 kg (Liu et al. 2018; Zhang et al. 2021; Cao et al. 2021; Wang et al. 2022).

Because of the complex nature of soils, different equipment is applied for the sampling. This usually includes excavation equipment, such as shovels, buckets, spades or spatula, augers, and core samplers (Piehl et al. 2018; Du et al. 2020; Katsumi et al. 2020). To avoid contamination, usually, the collected soil samples are stored in aluminum foil and bags, or cloth bags (Huang et al. 2019; Yu et al. 2021;). Other equipment used for site identification and measurement include GPS, ruler, measuring tapes, and calipers (Yu et al. 2021; Cao et al. 2021).

In this study, I adopted systematic quadrat sampling to collect the soil samples at sampling points. However, regarding the depth, I collected the soil samples from depths of 0-20 cm and 2-40 cm in the agricultural soils, while a 4-meter depth was adopted for the soil profiles. To harmonize the soil samples, composite sampling from 4 points was conducted. Finally, 500 g of soil samples were collected and transferred to the laboratory for analysis. For soil excavation, I used a hand auger. A shovel, bucket, aluminum foil, bags, pen, and measuring tape were also used for measurement, sample storage, and soil mixture.

2.8.3 Macroplastic purification in agricultural soils.

A limited number of methods were adopted for macroplastic purification in agricultural soils. Meng et al. (2020) and Stefano and Pleissner (2022) purified the macroplastic by soaking the contaminants in a container of tap water for 1- 2 hours. Huang et al. (2020) purified macroplastics by submerging them into 30% H_2O_2 for 72 hours at a temperature of 50^{o}C.The reasons for this deep cleaning in the Huang et al. (2020) study is probably because they used a limited number of samples, and the macroplastics were well mixed and heavily contaminated with the soil and organic matter content because they originated from sub-surface mulching. Other studies directly analyzed the macroplastic without putting it into any cleaning process (Kundu et al. 2021; Pietz et al. 2021).

Therefore, during my work, I adopted the method developed by Meng et al. (2020). I selected this method, because the method was used by other studies, and it successfully removes the dirt on the macroplastics. Additionally, unlike mulching macroplastics which were buried in the deep soils, the greenhouse macroplastics are usually on the surface without much dirt attached.

2.8.4 Methods of microplastic extractions in agricultural soils

The increasing threats and level of pollution by microplastic in the environment deserve efficient methods for the separation and detection of microplastic particles (Grause et al. 2022). In recent years, several methods have been developed to detect microplastic in the soil (Zhang et al., 2018). The pioneering studies of microplastic pollution in the terrestrial environment have embraced the analytical method used in aquatic studies without being conscious of the differences and the complex nature of the soil ecosystem (Thomas et al. 2020). The system of separation is more complicated in soils compared to those in water (Zhang et al. 2021). Further, even in the soil environment, agricultural soils have more separation complexity because of the high content of soil organic matter (SOM) (Li et al.

2019). Despite the presence of microplastic in the soil's environment, there is no globally accepted method for microplastic extraction (He et al. 2018; Li et al. 2019). Below is the steps used for microplastic extraction from soils.

a) Drying and sieving

Soil from agricultural farmland is usually processed before analyzing and extracting microplastic and macroplastic materials. Drying is imperative and typically performed to get samples of water-free reference and improve the precision of soil sample measurements, determination of the dry weight, and breaking the cell wall and membrane to increase aggregate dispersion and digestion efficiency (Thomas et al. 2020; Enders et al. 2020). Due to the lack of a standard method of microplastic extraction, the drying procedure is still contrasting in microplastic analysis. As in other environmental and analytical studies, soil can be dried via oven drying and surface drying in microplastic studies. Drying time depends on the moisture content of the samples and the set temperature, which is usually recommended to be ≤40 °C to prevent damage and alterations to the plastic particles (Li et al. 2019; Thomas et al. 2020). Whereas, Choi et al. (2020) used 60 °C for 48 h during the extraction from agricultural soils and other land uses in Korea. The highest temperature was applied by Liu et al. (2018) who selected 70°C for 24 h. Zhang et al. (2018) dried the samples using 25°C for many days until the samples dried. Moreover, Li et al. (2019) dried the agricultural soils using 40 °C for 24 hours. However, the main advantage of the oven-dry method is the ability to control environmental and atmospheric contamination (such as suspended fiber). The common disadvantage of this method is when the temperature exceeds the polymer limit, there is a high chance of alteration and destruction of the polymer's physical and structural properties (Contaminated Land Management Guidelines, 2011; Li et al. 2019; Wu et al. 2020).

Furthermore, soil sample drying could still be achieved by spreading and air drying the soil samples. For example, many studies such as Rezaei et al. (2019), Huang et al. (2020), and Cheng et al. (2020) adopted the same method by air-drying the samples. One of the advantages of air-dry is the ability to preserve the polymer's physical and structural properties. The main disadvantage is the difficulty in controlling environmental and atmospheric contamination (Thomas et al. 2020). The other method of drying the samples for microplastic studies is dry freezing. This is performed by subjecting the samples to a freeze dryer at a temperature of -20°C until the sample is completely dry (Enders et al. 2020). Dry freezing increases soil aggregate dispersion and reduces atmospheric contamination and sample loss due to overheating. On the other hand, low temperature increases plastic brittleness and fragmentation. Also, it is time-consuming, and the size of the freeze dryer makes it unsuitable for large samples.

After drying, the sieving is the next important step for plastic contamination analysis of soils. The sieve size depends on the definition of microplastic (≤5 mm) and the objective of the study. For the sieving of microplastic analysis samples, sieving through 1, 2, and 5 mm in compliance with standard mesh sizes and research objectives is recommended (Thomas et al. 2020). Most of the microplastic studies that concerned both larger and smaller microplastics sieved the samples using a 5mm sieve (Braun et al. 2020; Cao et al. 2021; Wu et al. 2022). Studies that deal with smaller microplastics and nanoplastic sieved the sample using a 2 mm sieve (Schorthorst et al. 2021; Zhang et al. 2022; Meng et al. 2020). Other studies that aimed at larger microplastic extraction adopt a 10 mm sieve (Zhang et al. 2018).

However, wet sieving could be also conducted when large sample volumes are collected, and drying is not required. The samples are processed directly using water to reduce sample size and discard particle sizes that are not required in the analysis (Piehl et al. 2018; Harms et al. 2021). This method is free from the destruction of the sample. However, a substantial number of samples may be lost. Also, the method is prone to environmental contamination especially since microplastic was reported in tap water (Kim and Lee, 2020).

The quantity of soil samples used for the laboratory analysis of microplastics differs by study. This may be caused due to differences in the study objectives and the level of microplastic contamination. Both small and gigantic quantities of samples were reported to be used by previous researchers. Some studies adopted the laboratory analysis by considering 1g, 3g, and 5g respectively (Wu et al. 2021; Beriot et al. 2021; Du et al. 2019). While many studies used 10g of soil samples (Zhang et al. 2018; Meng et al. 2020; Cao et al. 2021). Similarly, 20g and 50g of soil samples were adopted by other studies (Huang et al. 2019, Li et al. 2019; Steinmetz et al. 2022). The reasons for adopting a small sample size include economic reasons as well as it equally provides a high recovery rate (>90%) (Saadu and Farsang, 2021). However, other studies approached the laboratory sampling by considering plentiful soil samples (50-150g) (Li et al. 2019; Yu et al. 2021; Isari et al. 2021).

Therefore, during my research, I adopted the oven-dry method at the temperature of 40°C for 5 days because of the soil moisture. This was chosen because many previous studies adopted the same method and it properly controls the high rate of environmental pollution. Regarding sieving, I used a 5mm sieve throughout my research because the objectives of this study concerned all microplastics of < 5mm size. However, a 10 g size sample was used for the laboratory analysis. I selected this amount because it has been used in several previous studies (Meng et al. 2020; Cao et al. 2021). Importantly, a part of being economical, a high recovery rate of >90% was confirmed using a 10 g size (Saadu and Farsang, 2021).

b) Soil aggregate dispersion

Plastic contaminants are sometimes attached to the soil matrix to such an extent that their removal is only possible via the dispersion of soil particles. Such dispersion can be achieved using various techniques, but the application of dispersion methods in microplastic extraction studies is challenging because the process can change the plastic materials' form, shape, and size. Different ways for aggregate dispersions were reported by Thomas et al. (2020). A simple method in which tap water is added was reported to create dispersion (Piehl et al. 2018; Katsumi et al. 2020). Similarly, distilled water was used for this purpose and created a good result (Beriot et al. 2021; Lu et al. 2021; Schorthorst et al. 2021). The use of dispersion agent chemicals, such as an aqueous sodium hexametaphosphate solution, can have similar effects (Büks and Kaupenjohann, 2020; Steinmetz et al. 2022). Dispersion can also be attained by shaking the solution in orbital shakers (Li et al. 2019), or ultrasonication and pressurized mobile-phase leaching can effectively destroy soil aggregates and create soil dispersion (Yang et al. 2021; Wu et al. 2021). Additionally, mild grinding using a ceramic mortar and pestle, which does not destroy the plastic particles, can also be used to break soil clods (Li et al. 2019; Saadu and Farsang, 2021; Kumar and Sheela. 2021).

Consequently, I adopted the soil aggregate dispersion by using both ceramic mortar and pestle and an orbital shaker. These methods have been chosen over other methods because they have been used by many researchers of a similar nature and yielded promising results. Also, the method was reported to properly break the soil clods without being detrimental to plastic particles.

c) Digestion/Soil organic matter removal

Soil management, such as amendment with organic fertilizers, leads to excess SOM content in agricultural soils. Organic matter is higher in agricultural soils than in most natural sediments (Li et al. 2019). The SOM in agricultural soils is problematic in microplastic contamination studies because it has a similar density ($1\text{-}1.4 g/cm^3$) to most plastic polymers (He at al. 2018). Thus, digestion is performed to separate soils from microplastics because the presence of SOM interferes with the results of the microplastic analysis (Thomas et al. 2020).

Different studies have shown different ways to remove SOM from soils. Many studies approached SOM digestion using acidic methods. The acidic compounds used for digestion purposes include HCl, HNO_3, and H_2SO_4 (Bläsing and Amelung, 2018; Scheurer and Bikalge, 2018). However, numerous polymer types can be easily degraded and cause microplastic loss due to the high concentration of acid (Wu et al. 2020).

The second and one of the important SOM removal is through oxidative digestion. The main oxidative digestion compound used by different researchers in microplastic studies is 30% H_2O_2 (Li et al. 2019; He et al. 2018). Moreover, heating that ranges from $40–70^0$C for a few hours is required to assist digestion (Thomas et al. 2020). The 30% H_2O_2 is widely used because it has been successfully tested and applied to soils and sediments in previous studies. Also, it is one of the compounds with the highest recovery rate of > 98% (Hurley et al. 2018). Previous studies reported the main disadvantage of H_2O_2 application includes the changes in size and color of PE, PVC, PP, PA, PET, PC, and PU after 7 days (Nuelle et al 2014). Similarly, the second most important and most frequently used oxidative digestion is Fenton's reagent. The application of Fenton's reagent in the removal of SOM was tested by previous studies. While Vermeiren et al. (2020) reported a 90% recovery rate, Grause et al. (2022) recovered 94% of the microplastic in the soil. Similarly, Thomas et al. (2020) reported that some studies had a higher recovery rate of 100±10% removal efficiency using Fenton's reagent. He et al. (2018) believed that using Fenton's reagent has no destructive effect on polymers. In some cases, Fenton's reagent was combined with other SOM digestion compounds and reported very promising results (He et al. 2018; Li et al. 2019; Saadu and

Farsang, 2022).

Furthermore, SOM digestion is achieved using the alkaline method. This involves the use of alkaline compounds such as KAOH or NaOH. The alkaline solution breaks down the protein matrix and increases its solubility. Approximately 35-68% digestion of SOM was recorded while using the alkaline solution (Hurley et al. 2018). However, the alkaline solution was reported to degrade some plastic polymers and cause about 30% mass loss of PET and PC (Thomas et al. 2020).

Furthermore, SOM may be digested using enzymatic methods. This involves the use of enzymes (e.g., corolase; trypsin; proteinase; chitinase; lipase and amylase; protease; and cellulase) to selectively degrade the potential interfering matrix constituents without altering the microplastics (Thomas et al. 2020). Even though enzymatic digestion is efficient, the method is emerging and costly. It is time-consuming because it requires very long steps for sample preparation, and this may increase the rate of environmental contamination (Thomas et al. 2020).

d) Density separation

After the digestion of organic materials, density separation is an important step in microplastic analysis. This aim to separate the plastic materials from the inorganic compounds (e.g. silt, clay) of the soils, exploiting the buoyancy of the plastic materials on the surface as supernatants. It works on the principle that a high-density solution usually has a higher density than that of the average density of plastic materials. The minimum density of plastics in agricultural soils is 0.830 g/cm^3 (Poly(4-methyl-1-pentene)), whereas the maximum density is 2.15 g/cm^3 (Poly[tetrafluoroethylene-co-perfluoro(alkyl vinyl ether)]) (https://scipoly.com/density-of-polymers-by-density/). In density separation, the inorganic sediments settle down owing to their high density of 2.7 g/cm^3 (Wu et al. 2021), whereas microplastics and other materials (e.g., organic remnants) float on the surface.

Standardized density separation methods for microplastic analysis in the soil have not been developed. In addition, extraction procedures differ in terms of the technical setup, extraction duration, quality, and amount of salt added to the solution (Büks and Kaupenjohann, 2020). Different types of solvents and brine solutions have been used for density separation in the microplastic contamination including tap water, distilled water (Beriot et al. 2021), $NaCl_2$ (Yang et al. 2021), NaI (Li et al. 2019), ZnCl (Saadu and Farsang, 2022), NaBr, CaCl, and sodium heteropolytungstate solutions (Büks and Kaupenjohann, 2020). For low-density microplastic contaminants such as PE (0.92-0.97 g/cm^3), PP (0.90-0.91 g/cm^3), and PS (1.04-1.10 g/cm^3), saturated $NaCl_2$ (1.2 g/cm^3), normal water, and distilled water (1.0 g/cm^3) have been proven to be promising in separating the contaminants from the sediment (Zhang et al. 2018; Li et al. 2019; Meng et al 2020; Thomas et al. 2020). However, the low-density solvents are found to be very suitable in separating the low-density contaminants with over 90% recovery rates (Wu et al. 2020). Similarly, these floatation agents are the most commonly used ones, owing to their low prices, availability, and environmental friendly (Wu et al. 2021). On the other hand, separation by these floatation agents is not suitable for high-density microplastics such as PET (1.3–1.6 g/cm^3) and PVC (1.1–1.6 g/cm^3) because < 90% recovery rate is recorded. Also, samples need to be washed at least three times to obtain accurate purification and results (Wu et al. 2020).

High floatation densities such as ZnCl (1.5–1.7 g/cm^3), NaI (1.6–1.8 g/cm^3), NaBr (1.4–1.6), CaCl (1.3–1.5), and Na_2WO_4(> 3.0 g/cm^3) are suitable for the separation of both low and high-density microplastic polymers. The recovery rate of the floatation solvents in high-density polymers is >95% for many plastic polymers (Wu et al. 2020). When applied to the low-density polymer, a 100% recovery rate was achieved (Saadu and Farsang 2021). Importantly, only one wash was reported to extract both low and high-density microplastic contaminants (Wu et al. 2020). Some of the main challenges include their availability, associated costs, and environmental contamination. Li et al. (2019) compared three different salts (NaCl, ZnCl, and NaI) in the extraction of microplastic in agricultural soils and sewage sludge; they concluded that NaI and ZnCl are suitable for separating fibrous and high-density materials but have no beneficial effect on fragments and bulk microplastics. By contrast, these brine solutions have a high cost, low availability, and have harmful effects on the environment (Thomas et al. 2020; Wu et al. 2020).

Consequently, during my research, I adopted ZnCl as a floatation agent for density separation. I selected this solution because the other studies dealing with agricultural soils reported polymers with different densities (both low and high), shapes, and sizes from the sampled soils (Li et al. 2019; Wu et al. 2020). Similarly, ZnCl was commonly used in the density separation of agricultural soils because a high recovery rate of > 95% was reported in the previous studies (Li et al. 2019; Wu et al. 2020; Saadu and Farsang, 2021). Lastly, ZnCl is affordable compared to other brine solutions and it is easy to use as it requires no rigorous steps.

e) Filtration

The final step of the density separation of microplastics from soil sediments is filtration, the upper supernatant (including the microplastics) is collected and then it is filtered using a filter. Numerous scholars reported that different filter papers of different sizes were used for this purpose. Li et al. (2019) and Ragoobur et al. (2021) extracted microplastics using 0.45 μm filter papers. A higher size filter papers of 5-7 μm were also used by previous studies (Huang et al. 2020; Wang et al. 2022). Only a few studies extracted the microplastic using ≥20 μm filter papers (Isari et al. 2021). Further, 0.45 μm size filter papers were predominantly used in the previous studies (Corradini et al. 2019; Ding et al. 2020; Su et al. 2021). The reason for this dominance is that most of the studies were more concerned about the smaller microplastic contaminants.

Different filter types could be used because the selection of filter paper was proven to improve the quantity of microplastic contaminants (Li et al. 2019). The most common filter type used in the extraction of microplastic is nylon membrane (Li et al. 2019; Yang et al. 2021; Cao et al. 2021; Zhang et al. 2021b; Su et al. 2021). Li et al. (2019) compared different filter types of capacity and concluded that optimal microplastic extraction relies on passing the solution through nylon filters. The other most important filter papers include membrane cellulose nitrate filters (Wang et al. 2022; Ragoobur et al. 2021) and glass fiber filters (Huang et al 2020). Some studies filtrate the microplastic from ordinary filter papers (Ding et al. 2020; Meng et al. 2020; Isari et al. 2022). However, due to the small size of the filter papers as adopted by many studies, the filtration must be aided with a vacuum pump (Li et al. 2019; Meng et al. 2020; Cao et al. 2021).

Consequently, I adopted a combined method by using 20 μm nylon membrane filter papers for the first filtration and 0.45 μm Whatman filter papers for the second filtration. This was chosen to control the elimination of smaller microplastics during the counting because of a bigger microplastic cover-up. Besides, I selected nylon filter papers in my research because no debris or microplastic is lost as a result of washout through nylon filter papers. (Li et al. 2019).

f) Visual sorting

The identification and quantification of microplastics is arguably the most crucial process in microplastic studies (He et al. 2018). This involves the careful observation of microplastic characteristics and visual appearances such as size shape and color (He et al. 2018; Li et al. 2019; Thomas et al. 2020). Different methods have been used to visualize and identify microplastics.

The simplest method is the visual identification of plastic materials. The method is useful in the larger microplastic contaminants that can be seen with the naked eye on the surface of filters or any filtration medium (Grause et al. 2022). Visual identification methods have been used in many studies because they prove nondestructive properties to the samples, the method is cost-effective, and are easy to perform (He et al. 2018; Thomas et al. 2020). However, the main disadvantage of this method is its inaccuracy because it is associated with the overestimation of microplastic contaminants (He et al. 2018; Grause et al. 2022). For example, Eriksen et al. (2013) reported that 20% of contaminants initially reported as microplastics were later confirmed by other methods as aluminum silicate from coal ashes. Thomas et al. (2022) reported that the identification of microplastics using this method exhibits a 20-40% error. Similarly, about 32% of the smaller contaminants identified to be microplastics were later confirmed false by Raman technology (Lenz et al. 2015). Hence, visual identification should be combined with physical and chemical technology to give a reliable result (Lusher et al. 2017; He et al. 2018; Thomas et al. 2019).

g) Microscopy

Microscopic identification is another important method to identify and quantify smaller pieces. The knowledge of the physical characteristics of plastic is important in the microscopic analysis. Corradini et al. (2019) and Thomas et al. (2020) listed the most important properties to consider for identifying microplastics under a microscope. These are: geometric shapes, shiny surfaces, strong colors, and smooth and regular shapes. Similarly, the recommendation of MERI Guide (2017) further summarized the characteristics of microplastic materials under a microscope as no organic structure visible, uniform thickness of fiber materials throughout the length, and exhibition of homogenous color throughout.

Different types of microscopes have been used before in different studies. The most common microscopes used include scanning electron microscope (SEM) (He et al. 2018), stereomicroscope (Li et al. 2019; Harms et al. 2021; Fakour et al. 2021), and transmission electron microscope (Watteau et al. 2018). However, the characteristics used for microplastic identification can lead to errors and over-estimations of microplastic counts; thus, a sample of suspected plastic materials should be subjected to further examination via heat and a needle. Thus, during the "hot needle test" the needle is heated to attain a minimum temperature that changes the shape of the microplastics (Lusher et al. 2017; MERI Guide (2017). Similarly, Zhang et al. (2018) developed a new method by scanning the potential microplastic samples before and after heating and observing the visible changes in the microplastic under a microscope. Even though the method relied totally upon microscopic analysis, it may cause loss of sample due to overheating and sample melting. However, among all the microscopes used in microplastic studies, SEM and stereomicroscope microscopes are the most used. Their application over others may be because they are non-destructive and order of magnification at high resolutions.

h) Spectroscopy

Because of the complexity and homogenous nature of plastic contaminants with other contaminants, plastic materials are usually validated via further examination. The exact polymer type of the materials must always be confirmed after the plastic materials are visually identified. Numerous methods are used to characterize polymers, but Fourier-transform infrared (FTIR) and Raman spectroscopy are the most common methods considered in plastic contamination studies (Li et al. 2019, Braun et al. 2020; Li et al. 2020; Ragoobur et al. 2021). These techniques allow the simultaneous approach of identifying the physical and chemical properties of microplastics down to a resolution of 20 and 1 µm, respectively.

In FTIR spectroscopy, an interference wave interacts with the sample in contrast to a dispersive instrument, and the interacting energy assumes a well-defined wavelength range (Faix, 1992). Each molecule or chemical structure produces a unique spectral fingerprint, making FTIR analysis an excellent tool for chemical identification. FTIR spectroscopy is currently the most popular approach for characterizing microplastics. Attenuated total reflectance (ATR)-FTIR and microscopy-coupled FTIR can be used to measure particles with sizes of 20 mm to >20 µm, depending on the composition and molecular structure of the substance. FTIR techniques have been applied to analyze the agricultural soils of various regions (Piehl et al. 2018; Gies et al. 2018; Zhang et al. 2018). In the review of Veerasingam et al. (2020), ATR-FTIR was found to have been used in 60% of studies to analyze samples and characterize different polymer types for various environmental matrices. Despite the numerous advantages, the FTIR technique is not appropriate for analyzing samples with sizes for measuring the polymer composition of small samples. Hence the method has limitations in the heterogenous samples.

Raman spectroscopy is another sensitive method used to characterize polymers. It determines the vibrational modes of molecules and finds wide applications in the detection of inorganic and organic components in soils (Das and Guo, 2023). The vibrational data acquired in a Raman spectrum is rich in content about both the chemical and morphological structure of the polymer (Schlotter, 1989). After the samples are subjected to Raman rays, Raman spectra are usually generated and compared with a library to identify the exact composition at a certain percentage.

In this study, I adopted the Raman technique because it has been tested by previous studies (Braun et al. 2020; Li et al. 2020). Also, it proved non-destructive to the samples, and it can be used for measuring the small-size samples (Schlotter, 1989).

3. THE STUDY AREAS

The study was performed on abandoned greenhouse farmlands in Southeastern Hungary. The focus of this chapter is to provide information about the physical settings and the history of greenhouse farming in the area.

3.1 Site Location and History

Sampling was done on three abandoned greenhouse farmlands. The first area is located next to the city of Szeged (N 46.28990, E 20.18043); north of it, a site at Szentes was studied (N 46.5150, E 20.3325), and the third area was sampled further NE at Szarvas (N 46.3907, E 20.1526) (Fig.2). At Szeged, the farmland has the size of 1.52 ha. The greenhouses were built in the 1990s, and tomato was cultivated before they were abandoned in 2015. At Szentes the farmland has a size of 0.12 ha. Here the greenhouses were established in the 1990s and were used for pepper seed production before they were abandoned in 2011. In Szarvas, the farmland has a size of 1.75 ha. The greenhouses were established in the 1990s and have been abandoned since 2017. This site was used to produce tomato, pepper, lettuce, and cucumber. The studied greenhouse farmlands were probably abandoned because of economic factors.

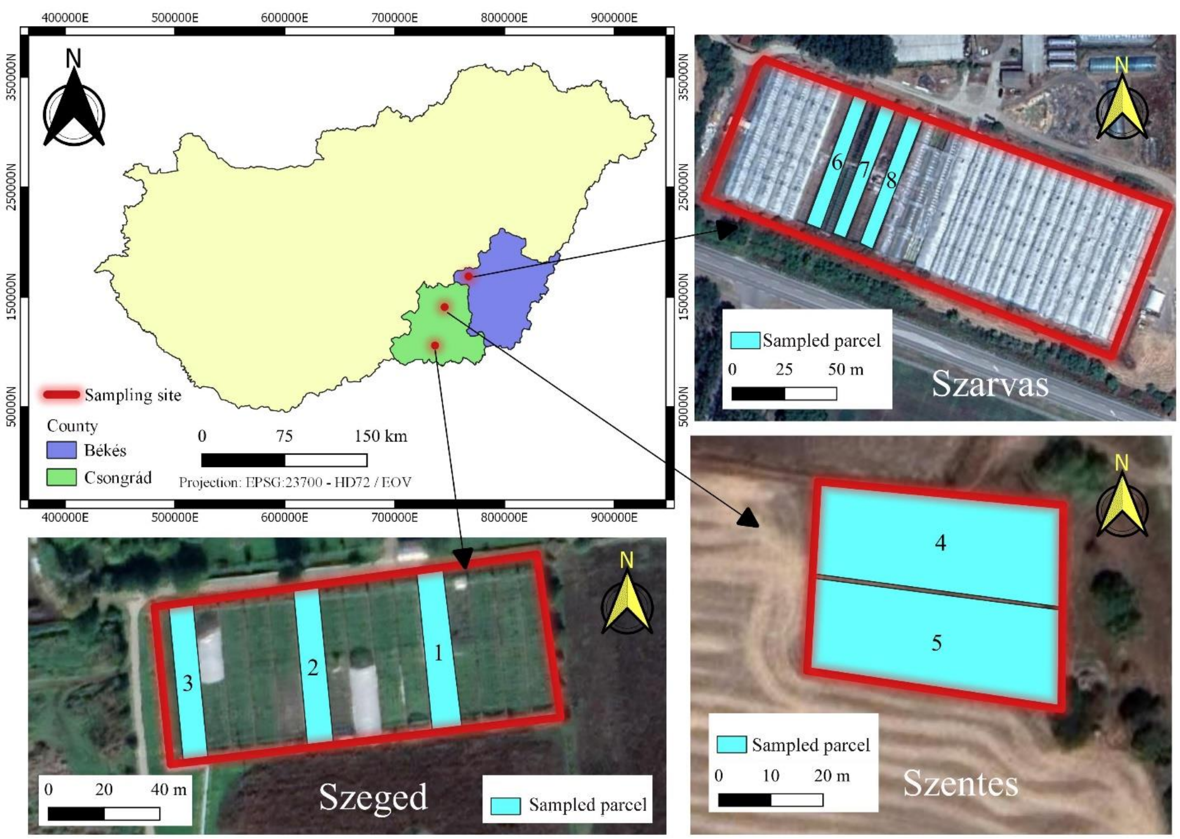

Figure 2. The study was performed on abandoned plastic greenhouse parcels in three areas in SE Hungary

3.2 History of Greenhouse Farming Activities

Greenhouse farming accounts for 5-6% of the total vegetable production area in Hungary, producing one-quarter of the vegetables harvested each year (Németh and Ehret-Berczi, 2014). The fruit and vegetable export of Hungary reached 1128 million Euros in 2022 (Statista, 2023), and a considerable amount of these fruits and vegetables was produced in greenhouses.

Plastic and glass greenhouses covered a total area of 697 ha in 2011 in Hungary (Mugnozza et al. 2011; Németh and Ehret-Berczi, 2014), which increased to 778 ha by 2021 (KSH, 2023). The area of greenhouses started to grow rapidly after 2012 but remained almost at the same level since 2015 (Fig. 3a). This study was performed in Békés and Csongrád Counties (SE Hungary), where the total area covered by greenhouses is relatively low on a country scale (Fig 3b) as the climate conditions therein are the most favorable for vegetable farming within entire Hungary. These climate conditions are probably also responsible for the gradual decrease in the areas covered by greenhouses since 2015 in these counties, especially in Csongrád (where the number of sunny hours is the highest). In southeastern Hungary, plastic and glass greenhouse farming are very popular because of the horticultural traditions of the area and the existence of thermal waters to heat the greenhouses.

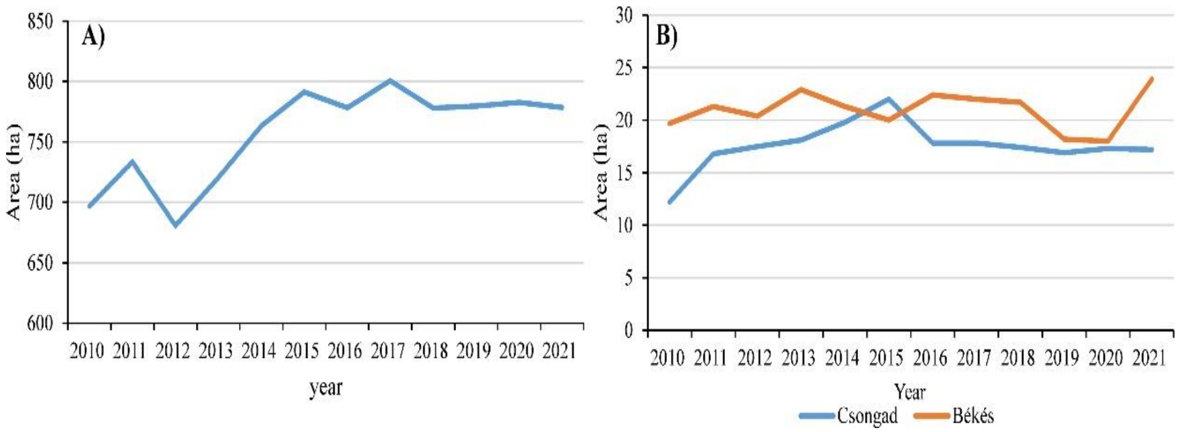

Fig. 3 Total areas covered by glass and plastic greenhouses in Hungary (A) and in Csongrád and Békés Counties (B). (Data source: KSH, 2023)

3.3 Climate

The climate of Hungary is affected by moisture-filled oceanic air masses and drier continental air masses. The climate of the country can be classified by Koppen's classification as a Cfa warm-temperate climate zone. Similarly, according to Trewartha's classification, Hungary is under the continental climate type characterized by a long warm season. At the national level, Hungary can be classified into a warm-dry climate category.

At the local level, the climate of the study sites is warm and dry (Dövényi, 2010; Szolnoki et al., 2013). In this study, I concentrated only on four climate elements (i.e. temperature, precipitation, solar radiation, and wind) and atmospheric contaminants that are directly related to agriculture and plastic degradation.

3.4 Temperature

The mean annual temperature ranges between 11 and 13 °C in most of Hungary. Comparably, the mean annual temperature at Szeged, which is the southernmost study site, is 14°C, which decreased to 13°C toward N and NE (Szentes and Szarvas). Based on measurements between 1991 and 2020, January is the coldest month of the year, but in any year, any winter month can be the coldest (Figure 4). The warmest period of the year is the end of July and the beginning of August. The average highest summer temperature over many years is 34°C in all the study areas; but the average lowest temperature is -17°C in the southern part of the area (Szeged and Szentes) which decreases to -18°C going further north of the study area (Szarvas). The weather in summer is more balanced, and the variability of summer temperatures from year to year is generally lower than in winter months.

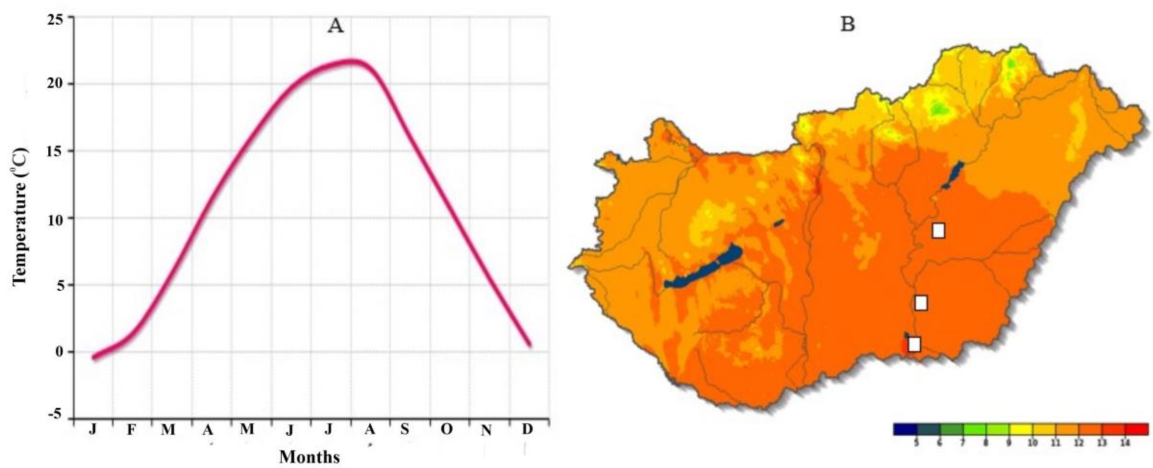

Figure 4: *Monthly mean temperatures in Hungary based on the 1991–2020 period.* (Data source: www.met.hu/en/eghajlat/magyarorszag_eghajlata/, 2023)

3.5 Precipitation

Precipitation is a highly variable meteorological parameter in Hungary, exhibiting significant inter-annual fluctuations. Most of the precipitation falls between May and July, while the driest months are between January and March (*Figure 5*). Previously, in normal years, February was the driest month, but recently January is the driest, and September and October have become significantly wetter, causing the disappearance of the secondary precipitation maximum in November. Unlike the southwestern region of the country where the highest annual precipitation is 800 mm, the study areas are among the driest regions with annual precipitation of 350–450 mm at all sites. The annual average snow cover was 28-30 days with a maximum thickness of 16-18 cm in Szeged. The duration of snow days increases to 30-32 days with a maximum thickness of 17 cm in Szentes. Moving further north, the number of days increases to 31-34 days with a maximum thickness of 17 cm (Dövényi, 2010).

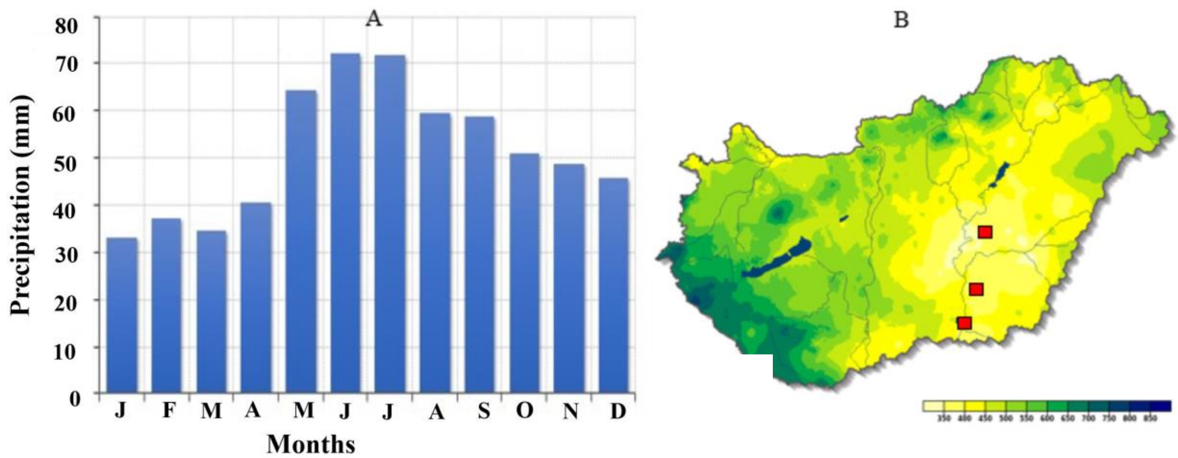

Figure 5. Average monthly precipitation in Hungary based on data between 1991 and 2020 (Data source: www.met.hu/en/eghajlat/magyarorszag_eghajlata/, 2023)

3.6 Solar radiation

The southern part of the Hungarian Great Plain is a region where the highest solar radiation values were observed in Hungary. In these regions, the global radiation ranges from 4800 to 5000 MJ/m^2, exceeding the mean of the other parts of the country (4600 MJ/m^2). Locally, the annual sunshine hours were 2020–2040 h/year in Szeged, which slightly decreased toward the north to 2000–2020 h/year for Szentes and Szarvas. Over the years, around 810 h/summer period of sunlight was recorded in the summer in Szeged and Szarvas. The value increased to 820 h/summer period in Szentes. Comparably, in the winter, the number of sunshine drastically reduced to 190 h/winter period in all the areas (Dövényi, 2010). The meteorological records from 1991 to 2020 show that most of the solar radiation falls between March and August (Figure 6). The highest solar radiation was recorded in July (680 MJ/m^2) while the lowest solar radiations were recorded in December (95 MJ/m^2).

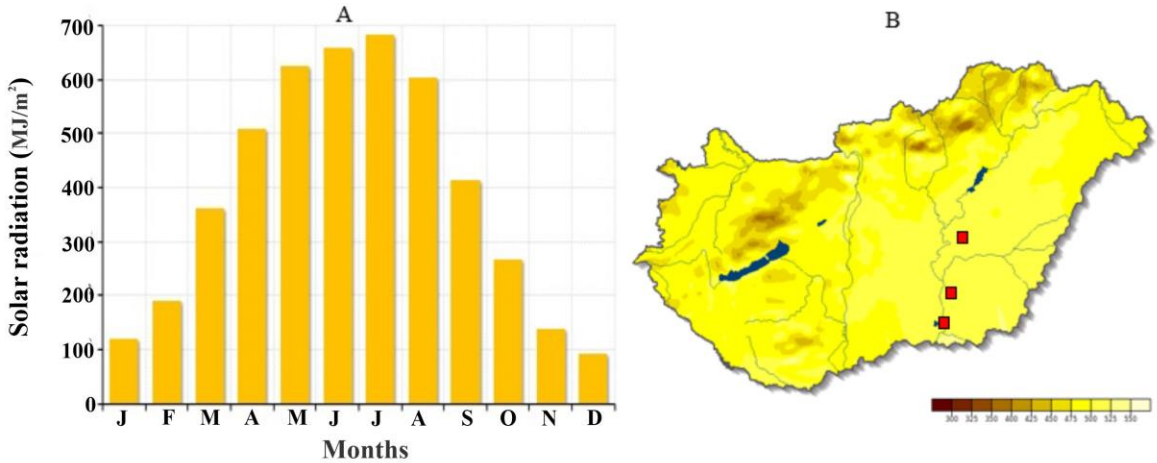

Figure 6. Average monthly global radiation in Hungary 2001-2020. (Data source: www.met.hu/en/eghajlat/magyarorszag_eghajlata/, 2023)

3.7 Wind

Hungary can be classified as a moderately windy region, as the average wind speed is 2-4 m/s. The dominant wind direction in the Great Hungarian Plain is northeastern[1]. Wind speed exhibits a characteristic yearly cycle, as the windiest period is the first half of spring and the lowest wind speeds are generally observed at the end of summer and the beginning of autumn (*Figure 7*). On average, there are 131 windy days per year in Hungary (i.e., when the speed of the strongest gust exceeds or reaches 10 m/s), and out of these, 33 days are stormy (i.e., with gusts exceeding 15 m/s). Locally, the wind direction in Szeged is NW and SE; NE and SE in Szentes while NE direction dominated the Szarvas area. Meanwhile, the maximum average wind speed is 3 m/s in all the areas (Dövényi, 2010).

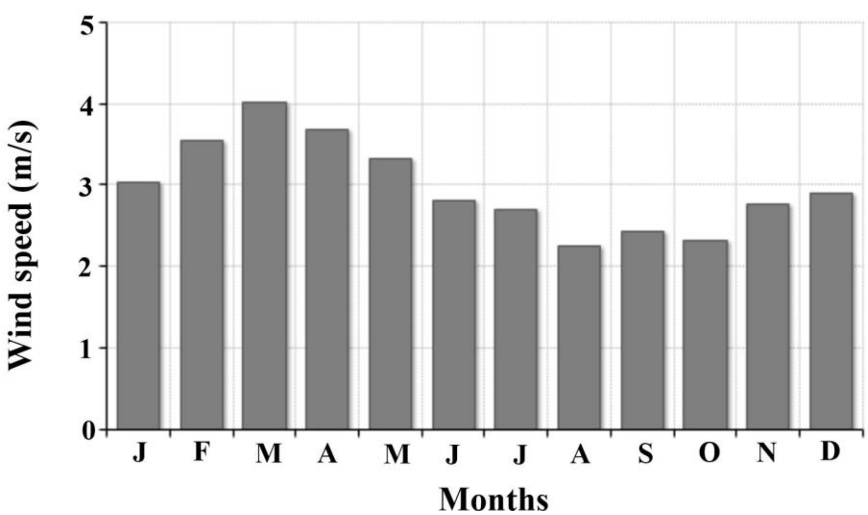

Figure 7: Average monthly wind speed in Hungary 2001-2020. (Data source: www.met.hu/en/eghajlat/magyarorszag_eghajlata/, 2023)

3.8 Atmospheric Contaminants

The major atmospheric pollutants that affect greenhouse plastic film are nitrogen oxide (NO_2) and sulfur dioxide (SO_2). The Hungarian air quality index shows that the country is covered with good and excellent air except for some pocket places in Budapest, Northeast regions (Miskolc and Sajozsentpeter), and the southwestern part of the country (Ajka and Varpalota) where the air quality index falls in the moderate and poor categories (https://legszennyezettseg.met.hu/en/air-quality/measurement-data/automatic-network).
Like most of the areas in Hungary, the study areas fall in the good category for both atmospheric contaminants (NO_2; 40-90 µg/m³ and SO_2; 100-120 µg/m³). Specifically, in Szeged in 2022, the annual average of NO_2 and SO_2 were 13.44 µg/m³ and 13.11 µg/m³ respectively (Figure 8). The highest contamination of NO_2 was in March with a mean monthly contamination of 22.53 µg/m³ while the lowest contamination was recorded in June with a mean monthly contamination of 8.42 µg/m³. Similarly, the highest contamination of SO_2 was recorded in December with a mean monthly of 14.8 µg/m³ while the lowest contamination was recorded in January with 11. 12 µg/m³.

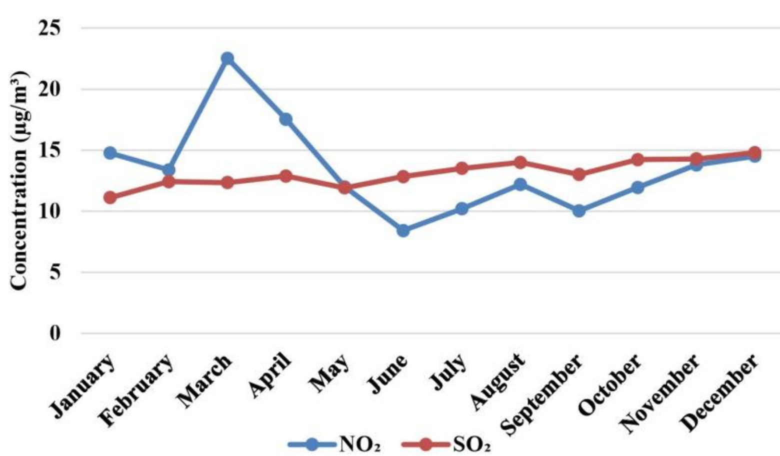

Figure 8: Average monthly contamination of NO_2 and SO_2 in Szeged 2022. (Data source: https://legszennyezettseg.met.hu/en/air-quality/measurement-data/automatic-network, 2023)

3.9 Soils

The study sites at Szeged and Szentes are located on a plain area with Phaeozem soil developed on loess (Dövényi, 2010; Szolnoki et al. 2013). The mechanical composition of this soil is clay loam and clay which do not contain carbonated lime[1] They are strongly acidic in some places.

The natural soil type in Szarvas is Chernozem developed on infusion loess (Dövényi, 2010). The top layer is rich in humus content, as it contains 3-4% humus. Slightly lighter colored subsoil containing less humus (B level) is found in the 30-60 cm.

[1] https://portal.nebih.gov.hu/-/magyarorszag-talajtipusai

4. MATERIALS AND METHODS

4.1. Sampling

Sampling was performed on rectangular parcels used as 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) (Figure 2). Systematic random sampling was used in the greenhouse farms, three parcels were selected at Szeged out of 15 and out of 30 plots in Szarvas. In Szentes, the abandoned parcels were not part of a greenhouse field; thus, both parcels were selected with similar sizes.

For macroplastic contaminants, two observers picked and collected all visible macroplastic debris on the surface of the entire selected parcels (Figures 9 and 10). Sample breakage was tried to be avoided, but there was minimal breakage. All collected plastic particles were precleaned to remove the attached soil by scrubbing their surfaces. The materials were later stored in large containers, and they were transferred to a laboratory for further analysis.

Figure 9. An observer picked visible macroplastic in an abandoned parcel.

Samples for microplastic analysis were collected from the greenhouse and control farmlands. For the control plot, four samples were collected at Szeged, 600 meters away from the greenhouse farmland. The control area is a ploughing farming area. For the greenhouse farmland samples, metallic auger, tape, hand shovels, and buckets were used for sample collection. The selected parcels were equally divided into two parts (Figure 10). 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 a half part of a plot. In this way altogether 32 soil samples were collected. The same method was used for control sampling and 4 samples were collected.

Thus, in total, 36 soil samples were collected from the upper and lower layers of the soil.

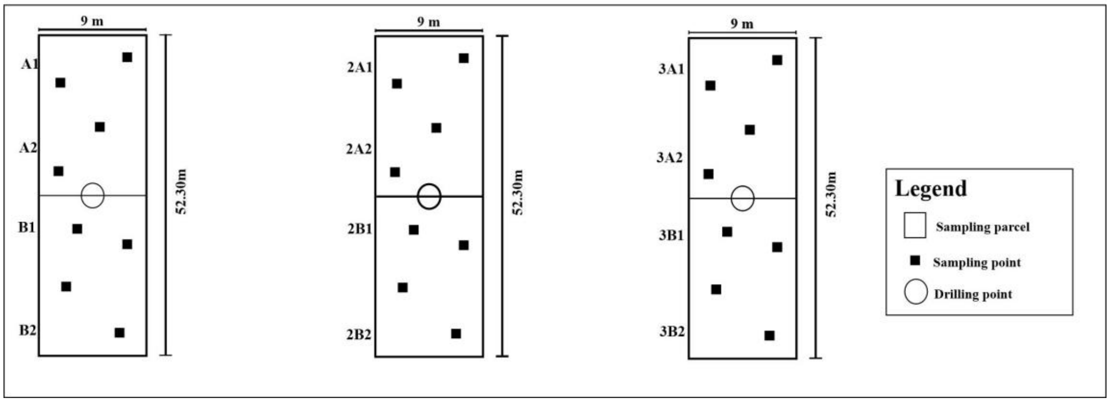

Figure 10. An observer picks visible macroplastic in an abandoned parcel.

Further, in the middle of each sampling plot drills were made to collect samples from the entire soil profile. Soil samples were collected at 20, 50, or 40 cm intervals down to the level of the groundwater (Figure 11). About 70 soil profile samples were collected in the study areas. The samples were wrapped with aluminum foil before being stored in bags and transferred to the laboratory for further analysis.

Figure 11. Soil profile from one drill.

4.2. Laboratory works

4.2.1. Preparation of macroplastic samples

This study adopted the method developed by Huang et al. (2020) with minor modifications (Figure 12). The collected macroplastics were submerged into 15-liter buckets filled with tap water and soaked for 48 hours to remove all impurities and attached soil particles. The plastics were rinsed thereafter. 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 using an electric analytical balance.

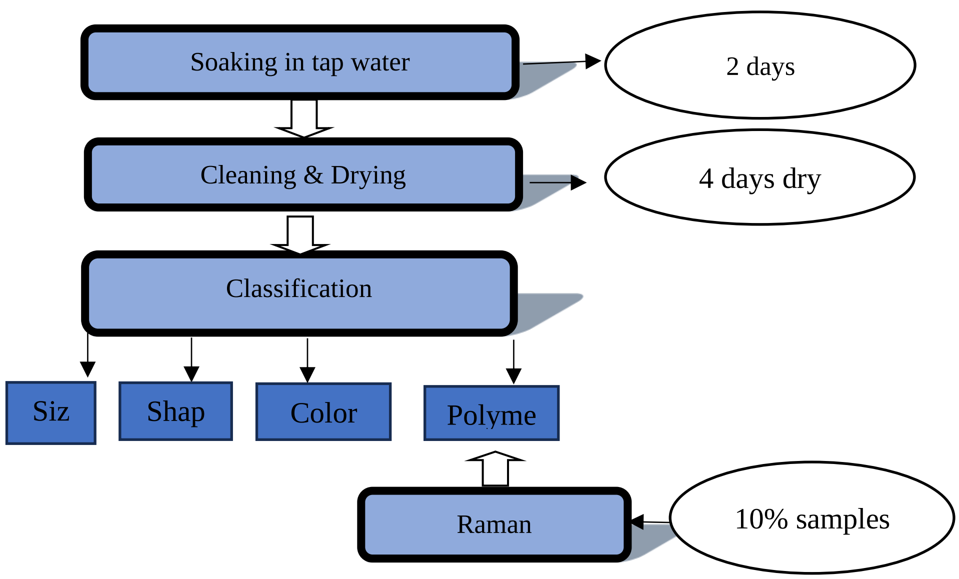

Figure 12. Schematic diagram of macroplastic purification method.

The size of the macroplastics was measured at the contaminants' longest axes for size categorization using a millimeter precision ruler. The macroplastic pieces were grouped into the following size classes: 0.5–1.0, 1–5, 5–10, 10–15, and >15 cm. These classes were further grouped into smaller macroplastic (≤5 cm) and larger macroplastic particles (≥5 cm).

The contaminants were also categorized by their color, such as transparent white, gray, blue, black, red, and green. Similarly, they were also grouped into shapes based on their physical appearance (e.g., film, fragment, and fiber). Films are the remnant of plastic foil used for greenhouse coverage. Fragments are broken pieces of hard plastic broken pipes, containers, and other related plastic materials. Fibers are threads that are used for tightening the greenhouse structure which are usually thick throughout their length variations (Figure 13).

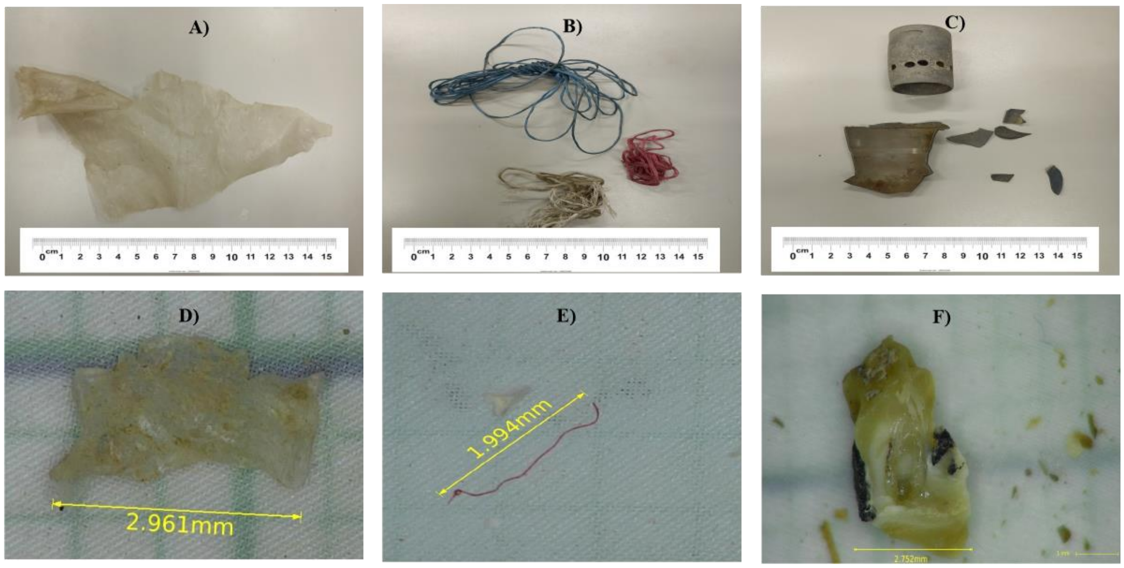

Figure 13. Some of the recovered macro-and microplastic contaminants. A-C are macroplastic (A) film, (B) fiber, (C) fragment. D-F are microplastic(D) film, (E) fiber, (F) fragment.

Approximately 10% of the macroplastic pieces were taken to a Raman spectroscopic analysis for polymer composition identification. Based on the results of the spectroscopy, they were classified as PE, PVC, PET, and PP.

The contaminants were finally categorized according to their probable origin. Agricultural contaminants were those directly related to agriculture and were the products of agricultural usage. These included aged plastic greenhouse films, fragments of broken irrigation pipes, stings, agrochemical bottles, and packages. In comparison, nonagricultural plastic contaminants probably had a communal waste origin, and they were not directly related to agriculture. These included candy and biscuit wrappers and disposable cups. Plastic equipment use was minimized throughout sampling and laboratory analysis to prevent contamination.

4.2.2. Testing various microplastic extraction methods

The validation test was carried out on soils obtained from two agricultural farmlands (greenhouse and plow land). Briefly, light-density (PE, PP) and heavy-density (PET, PVC, and PU) microplastics were both obtained from agricultural environments and were prepared by chopping the macroplastic materials into particles of <5 mm. The chopped plastics were incorporated into the soil and mixed thoroughly. Three replicates were made for each set.

Different salt solutions were tested to select the best result for this study. According to this recovery test, $ZnCl_2$ (1.5 g/cm3) and NaI (1.8 g/cm^3) were tested very well in extracting both high and low-density microplastic contaminants with over 80% recovery rate (Table 2). Further, they yielded good results in the extraction of contaminant structures such as fibers, film, and fragments. On the other hand, NaCl (1.2 g/cm^3) and distilled water (0.99 g/cm^3) were only tested well in the extraction of low-density contaminants. In terms of structure, they were reliable in extracting fibers and film structures.

Consequently, I chose $ZnCl_2$ over NaI, because $ZnCl_2$ yielded more PU microplastic (5 out of 10 pieces) compared to NaI which did not yield any PU contaminants. Likewise, the $ZnCl_2$ was chosen because macroplastic foams were recovered from the areas hence there is the possibility of their fragmentation to microplastic foams.

Table 2: Validation of the method results

S/n	Solution	Density (g/cm^3)	Light density		High density			Total	Total Recovery rate (%)
			PP Fiber	**LDPE Film**	**PET Fragment**	**PVC Fragment**	**PU Foam**		
1	$ZnCl_2$	1.5	10 (10)	10 (10)	10 (10)	10 (10)	10 (5)	50 (45)	90
2	NaI	1.8	10 (10)	10 (10)	10 (10)	10 (10)	10 (0)	50 (40)	80
3	NaCl	1.2	10 (10)	10 (7)	10 (4)	10 (0)	10 (0)	50 (21)	42
4	Dist. water	1	10 (10)	10 (9)	10 (0)	10 (0)	10 (0)	50 (19)	38

Note: Bracket values mean the number of pieces recovered

4.2.3. Preparation of microplastic samples

To evaluate microplastic contamination, a method developed by Li et al. (2019) was modified and used (Figure 14). The soil was oven-dried at 40°C, gently ground into smaller pieces, and sieved with a 5 mm sieve. Digestion of soil organic matter was performed through a mix of 10 g of soil, 40 ml of 30% H_2O_2, and 10 ml of Fenton reagent in the 250 ml conical flasks. The solution was heated at 70°C until all liquid evaporated. The containers were then immersed in cold water and a few drops of butyl alcohol were added to reduce the samples' spout out. Consequently, $ZnCl_2$ [1.5 g/cm^3 (5 mol/L)] was used as a flotation salt, and 40 ml of the solution was added. The complete solutions were capped with aluminum foil and shaken for 1 h at 200 rpm in an orbital shaker, after which they were emptied into 100 ml beakers and allowed to settle for 24 h. Approximately 20 ml of the upper supernatants were collected with a glass pipette and 20 ml of $ZnCl_2$ 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. These extracts were later 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.

The microplastic contaminants were finally categorized according to shapes such as films, fiber fragments, foam, and beads (Figure 13D-F). Similarly, they were also categorized into five different size classes: 0-1 mm, 1-2 mm, 2-3 mm, 3-4 mm, and 4-5 mm. Further, the contaminants were categorized based on their color appearance into transparent, blue, black, white, yellow, green, and red. However, the samples from the control site were put through the same methods as described above. Moreover, blank samples were created and used throughout the laboratory analysis. The atmospheric contaminants found in the blank samples were subtracted from the main samples.

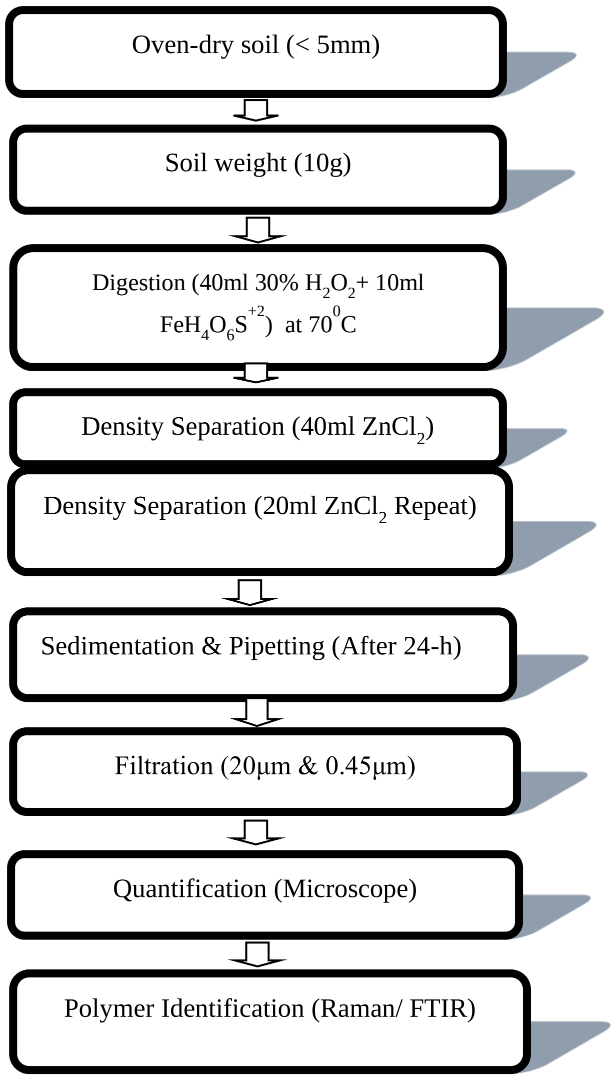

Figure 14. Schematic diagram of microplastic extraction method.

4.2.4 Visual sorting via microscopy and Spectroscopy

The filter papers with suspected microplastic debris were taken to the light microscope (Inspex II) for microplastic identification and quantification. The 50x magnification was used to zoom out the potential microscopes for proper identification using visible lights. Materials with strong colors, shiny surfaces, smooth sides, and geometrical shapes were considered plastic debris as described by MERI Guide (2017) and Corradini et al. (2019). Furthermore, 1-2 % of the suspected plastic particles were confirmed using a hot needle method as described by MERI Guide (2017). Subsequently, the larger microplastic particles were later taken to the Raman spectroscopic analysis for polymer identification.

4.2.5 Soil physical parameters

To reveal the pedological background of microplastic migration in the soil, the following physical parameters were measured: particle size distribution, saturated hydraulic conductivity, bulk density, and porosity.

For the particle size distribution, a Fritsch Analysette 22 MicroTec laser particle sizer was applied. This instrument uses two linearly polarized He-Ne lasers; a green (λ = 532 nm, p = 7 mW) and an infrared (λ = 940 nm, p = 9 mW) laser. The measuring range of the instrument is 0.08-2000 µm. The preparation of the samples was achieved by adopting the method of Kun et al. (2013) and Markovic et al. (2023); thus, a chemical dispersion agent was not applied to keep away from altering the effect of dispersion in the measurement (Markovic et al. 2023). Based on the diffraction patterns obtained on the sensor, I used the Fraunhofer method as described by Orsolya (2014). Further, I selected the first measurement result for the data of this study. Also, I used the Udden Wentworth scale for particle size categorization.

For soil bulk density, I used the weighing bottle method as described by the Directorate of Irrigation Research & Development (DIRD, 2009). The mass of the soil was determined by weighing the oven-dry soil sample. I determined the mass of the empty bottle, the mass of the bottle + soil, and the volume of filling the bottle. Then, the bulk density was calculated from the mass and volume of the soil using the following formula:

$$Bulk\ Density(\text{g/cm3}) = \frac{M2 - M1}{V}$$

Where *M1* is the mass of the empty bottle (g), *M2* is the mass of the bottle + soil (g), and *V* is the volume of water filling the bottle (cm^3).

For saturated hydraulic conductivity, I used permeameters method as described by DIRD (2009). Both the constant water head method and falling head method were used because the nature of the soil samples contained a high proportion of clay. The following formula was used for the constant water head method:

$$Saturated\ Hydraulic\ Conductivity\ (\text{cm/min}) = \frac{QL}{At(L + h)}$$

Where: D is the diameter of the permeameter (cm), Q is the volume of percolate collected ($cm^{3,}$), L is the length of the soil column (cm), A is the cross-sectional area of the permeameter (cm^2), t is the time for which percolate collected (min), and h is the depth of water above the soil (cm).

The following formula was used for the falling head method:

$$Saturated\ Hydraulic\ Conductivity\ (\text{cm/sec}) = \left(\frac{al}{At}\right)\left(\frac{h1}{h2}\right)$$

Where: *d* is the diameter of standpipe (cm), *a* is the cross-sectional area of standpipe (cm^2), *L* is the length of the sample (cm), *D* is the diameter of the sample (cm), *A* is the cross-sectional area of the sample (cm^2), *h1* is the initial hydraulic head (cm), *h2* is the final hydraulic head (cm) and *t* is the time taken for change in head (sec).

4.3 Statistical analysis and quality control

Both descriptive and inferential statistics were used in our analyses. Descriptive statistical analysis was performed using Microsoft Excel, whereas inferential analysis was conducted using SPSS (version 22). Differences in the number of microplastics among soil depths were determined using a simple Student t-test. The relationship between microplastics and soil depth was determined using Spearman's rank correlation. ANOVA was used to determine the relationships among soil profiles. A bare minimum of plastic materials was used during sampling and laboratory analysis. For the data presentation and analysis, a combination of mean and mean error was used (mean ± standard error). However, contamination prevention techniques, such as cleaning the auger before the next sampling in the field and avoiding sample mix, were strictly ensured in the field. Similarly, rinsing the apparatus with distilled water three times was adopted throughout the laboratory processes, during which researchers always wore a cotton lab coat and hand gloves. Aluminum foil was used from sampling until the final stages to cover the analyzed samples to prevent atmospheric contamination.

5. RESULTS AND DISCUSSION

Within this section, the results of the PhD research are presented, and the findings are discussed and compared with the related literature. At the end of the chapter, the implications of plastic contamination in agricultural areas are discussed.

5.1. Macroplastic abundance

Macroplastic debris was discovered in all sampled parcels of the greenhouse farmlands, where the overall mean abundance was 2655 pieces/ha. The maximum mean abundance, 4328 pieces/ha, was found in Szeged, and the second-highest abundance, 3513 pieces/ha, was found in Szarvas. Szentes had the lowest abundance, 125 pieces/ha, thus the plots at Szeged and Szarvas are 34 and 28 times more polluted. The overall mass average of macroplastic pollution was 5 kg/ha. The highest contamination was found in Szarvas (10.68 kg/ha), followed by Szeged (6.4 kg/ha) and Szentes (1.17 kg/ha); thus, the field at Szarvas and Szeged are 9 and 5 times more contaminated if one considers the weight of the pollution. Thus, Szeged fallow greenhouse sites are more macroplastic contaminated in terms of the number of macroplastic contaminants while Szarvas fallow greenhouse is more polluted in terms of weight. However, macroplastic is not evenly distributed in the greenhouse fields. For example, parcels No. 1 and 2 in Szeged had the highest record of macroplastic contamination, 4851 and 5592 pieces/ha respectively. This is followed by parcel No. 6 in Szarvas with 4360 pieces/ha. The least contamination was recorded in parcel No. 5 in Szentes with a total abundance of 66 pieces/ha in the parcel (Figure 15). Compared to these fallow greenhouses, no macroplastic contamination was found in the control site (at Szeged).

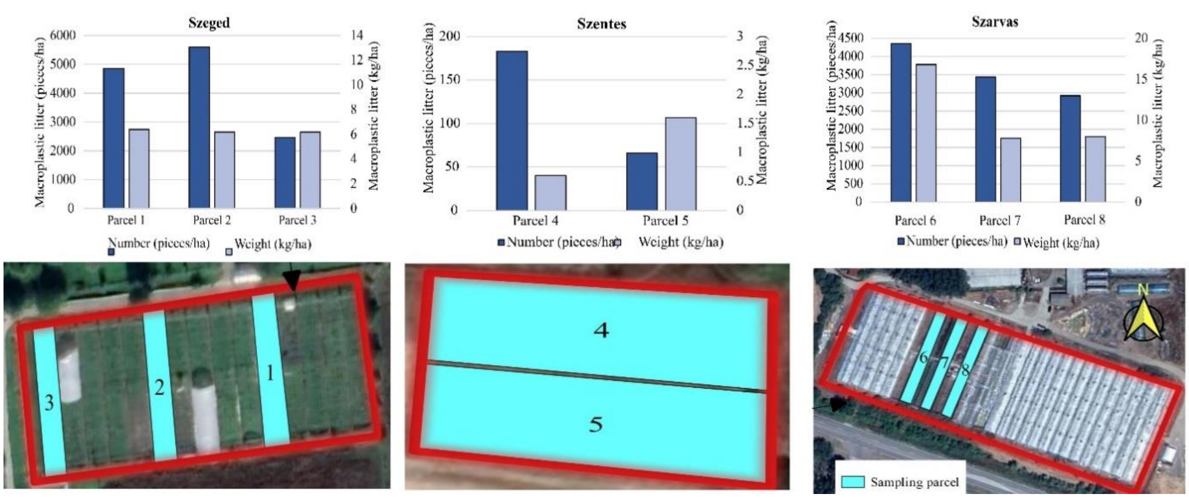

Figure 15. Total macroplastic abundance (in number and weight) at the studied parcels

Despite the few investigations on the presence of macroplastics in agricultural soils, this result matched the findings of the few studies that measured the amount of macroplastic contamination on agricultural surfaces. According to Piehl et al. (2018), the abundance of macroplastic litter (206 pieces/ha) in the agricultural soils of Germany was almost 13-fold less than in the study locations in Hungary. Similar contamination has been reported in Tanzania, where Kundu et al. (2021) discovered 3.3–36.6 kg/ha of macroplastics on agricultural soils, but Huang et al. (2020) found 83.6 kg/ha of macroplastics in Chinese mulching farmlands, indicating substantially higher contamination. The Hungarian sites in this study were significantly more polluted when compared to Kawecki and Nowack's (2021) findings, which indicated a median emission rate of macroplastics of 0.0006 to 0.06 kg/ha/year in Switzerland. However, compared to Stefano and Pleissner's (2022) findings, which showed a macroplastic contamination value of 9247 pieces/ha in arable land treated with compost in Germany, our data demonstrate significantly less contamination. The differences in the management of conventional agricultural fields and greenhouse farmlands may account for the differences in the amount of macroplastic in these studies.

The studied sites reflected high variation in macroplastic abundance. Differences in greenhouse farming duration, farmland abandonment duration (Liu et al. 2018; Meng et al. 2020), clearing activities, and climate may also be contributing factors to this variance. For instance, the extended duration of greenhouse farming in Szarvas, which lasted for 27 years, may be responsible for the high quantity of macroplastic pollutants in the area. In the meantime, a shorter period of greenhouse farming (21 years) may be associated with the low level of macroplastic pollutants in Szentes. Furthermore, the greenhouses at Szentes did not form a part of a larger greenhouse farm, preventing cross-contamination from nearby greenhouses from worsening the plots' macroplastic pollution. Additionally, the area at Szentes has been abandoned since 2011, whereas greenhouse farming was finished 4 and 6 years later in the other two sites (unfortunately, no information could be collected on the way of clearance of the foils, as in some cases the greenhouses are nicely cleared after the termination of the activity, while in other cases the greenhouse is just abandoned without proper clearance). The reasons for the abandonment include climatic conditions as well as economic factors. The fact that 10 times more macroplastic contamination was found in Szeged than in Szentes and twice as much in Szarvas could be explained by climate conditions as well. Szeged receives a higher amount of solar radiation (2020–2040 h/year), which is higher than that of other locations. The intensive solar and UV radiation accelerates the deterioration of greenhouse films according to various researchers (Dilara and Briassoulis, 2000; Vox et al. 2008; Babagyayou et al. 2020). Furthermore, other climatic factors, like water and wind have been shown to transport light macroplastic contamination to other areas (Kundu et al. 2021). Thus, it makes sense that an area with a long history of abandonment (eg., Szentes) has less macroplastic litter. Besides, long-term plastic greenhouse agricultural practices could contaminate the soil with macroplastics more, due to the gradual weathering of the foil. The outcome also suggested that prolonged abandonment might cause the contaminants to disperse to other locations.

However, no macroplastic materials were discovered in the control site of this study. This indicates that the macroplastics recovered in the greenhouse fallows were site-based contamination that occurred either as a result of greenhouse plastic materials fragmentation or anthropogenic factors. The unavailability of macroplastic in the control area can be traced because of the distance of the site from the greenhouse farmlands (600 m) or because the areas were separated by water canals that may stop the horizontal movement of loose plastic contamination due to the action of wind and water runoff. Another reason for our inability to discover larger plastic contamination may be because of the difference in land management as the control site is a ploughing site. During the fieldwork of the control site, the area was newly plowed, this may also determine the abundance as the macroplastic contamination might be buried in the soil depths.

5.1.1. Sizes and fragmentation of macroplastics

Five fragmentation categories were used to group the macroplastic sizes. The size class of 1-5 cm was the most prevalent (46%) in Szeged, whereas 15-10 cm was the least (5%) prevalent (Figure 16). In contrast, only two categories were noted in Szentes, where large particles (>15 cm: 80%) predominated over smaller ones (10–5 cm: 20%). The size class with the highest abundance in Szarvas was 1.0-0.5 cm (42%); other classes had relatively comparable distributions, but the class with the lowest abundance was 5-1 cm (12%).

On the other hand, the highest weight abundance class was >15 cm (73%), and the lowest weight abundance class was found to be 1.0-0.5 cm (0.07%) in Szeged. In comparison, just 1% of the measured macroplastics in Szentes were in the 10-5 cm class, while the majority of the macroplastics (99%) were larger than 15 cm. The classes of 5-1, 10-5, and 15-10 cm showed the highest abundances in Szarvas (70%) followed by the largest particle sizes (>15 cm). The class of objects 1.0-0.5 cm in diameter had the lowest abundance (0.07%).

The breakdown of macroplastics into microplastics is detectable when the abundance of the size classes is analyzed in detail in Figure 16. In Szeged, macroplastic fragmentation was visible in every parcel because there were more small 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 the Szarvas locations, and 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 microplastic pollutants were discovered on the soil surface.

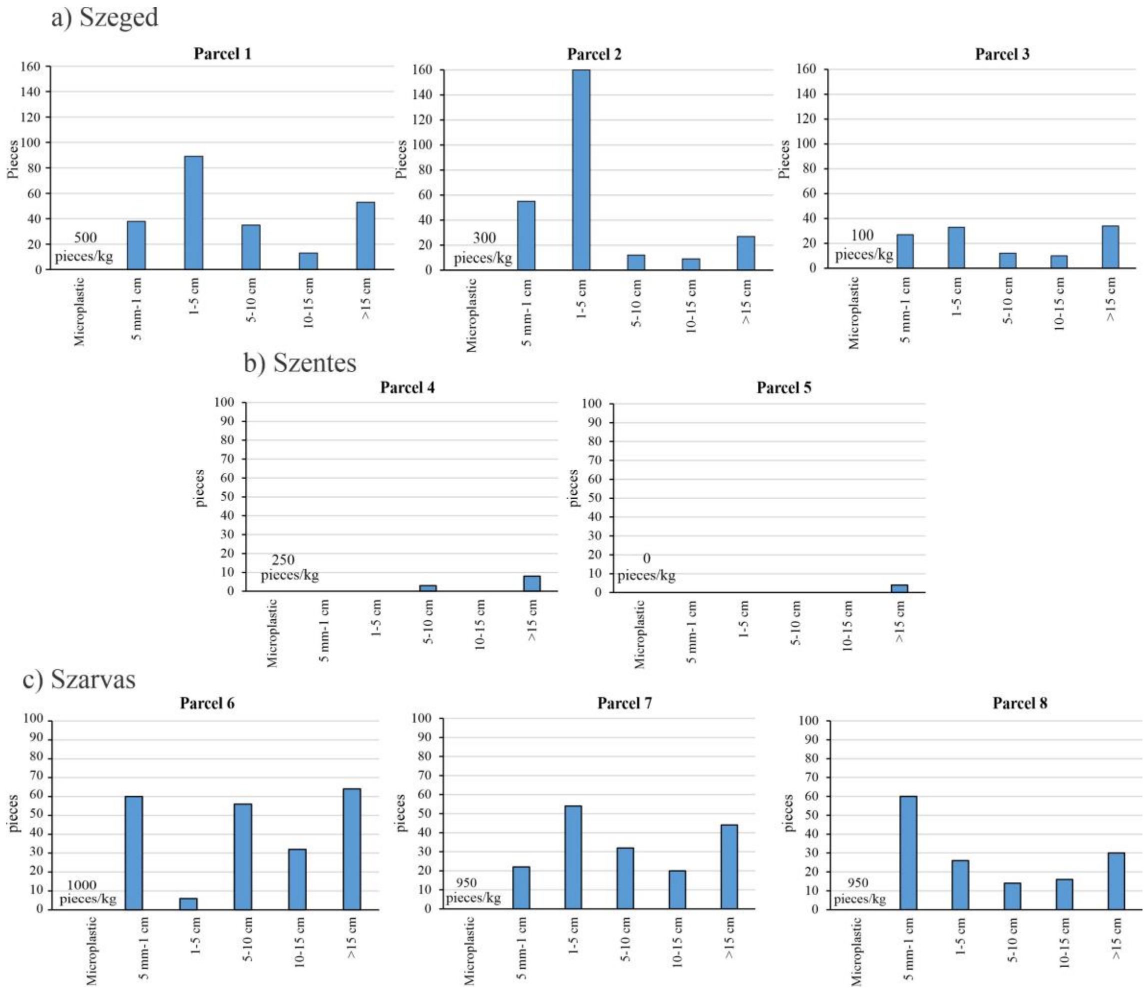

Figure 16. Macroplastics and microplastics collected from the surface of fallow greenhouse plots at three study areas (a) Szeged. (b) Szentes and (c) Szarvas.

This study found different sizes of macroplastics in the studied greenhouse farmlands. This result is consistent with other pertinent findings that showed plastic materials are broken into various sizes and categories when used as agriculture mulch (Liu et al. 2018; Huang et al. 2020; Meng et al. 2020). Similar reports of the dispersal of plastic pollution in traditional farmlands and along riverbanks have been made (Piehl et al. 2018; Kundu et al. 2021). Thus, the availability of macroplastics on the surface is influenced by the rate of litter fragmentation. For instance, in the case of Szeged, the climate and the atmospheric contaminants (NO_2 and SO_2) accelerated the quality degradation, stress, and aging of plastic materials, resulting in a higher concentration of contaminants than in other farmlands.

Furthermore, our results support the hypothesis that plastic film covers and other plastics used in greenhouses break down into smaller pieces. The gradual fragmentation process is brought on by a combination of climatic, agrochemical, atmospheric contaminants, structural, and environmental variables (Dilara and Briassoulis, 2000; Vox et al. 2008;

Julienne et al. 2019; Babagyayou et al. 2020). Plastic materials must be gradually fragmented over time, first visible cracks appear, then holes will be on the particle surface, and lastly, larger pieces are broken into smaller ones (Weinstein et al. 2016; Li et al. 2022). The degree of fragmentation occurred in the following order: Szarvas greenhouse > Szeged greenhouse > Szentes greenhouse. Similarly, microplastics were discovered in the soil surface of the investigated plots, suggesting a gradual and ongoing fragmentation and contamination process. This outcome is consistent with Li et al.'s (2020) observation that the majority of plastics used in mulch farming degrade and produce >2cm macroplastics. In contrast, there was less macroplastic and microplastic contamination in the Szentes greenhouse, most likely because plastic films and other plastic items were probably adequately removed when the plots were abandoned. Another factor could be the long-term horizontal displacement of macroplastic trash from the location to other points as a result of the greenhouse's abandonment.

High annual temperature (10.4°C–10.6°C), high seasonal temperature variations, high annual solar radiation (2000–2040 h/year), increasing frequency of heavy rainfall and hail, and thermal water heating of the greenhouses could all contribute to an increase in the advanced fragmentation states in our study areas. Due to its widespread use as a covering for plastic greenhouses, PE was the most readily available plastic material for fragmentation in the study plots. As a result, it experienced significant environmental stress from factors like solar radiation, precipitation, wind, and air pollution (Andrady, 2003). Likewise, the interior surfaces of PE foils were coated with the agrochemicals utilized in the greenhouses. Because PE has low densities (0.917-0.960 g/cm^3), low melting points (135°C), poor mechanical strengths, rigidities, and hardness, it was easily broken into smaller pieces of microplastic (Andrady, 2003). Given that PE has a simpler chemical structure than any other polymer, environmental factors like photooxidation, thermal oxidation, and chemical hydrolysis rapidly weaken the chemical chain structures of PE polymers and induce aging (Andrady, 2003).

5.1.2. Origin of macroplastic contaminants

The research revealed that not all macroplastics originated directly from agriculture (Figure 17). The overall abundance showed that only 90% of the litter originated from agricultural sources, and 10% of the litter had non-agricultural origins (e.g. candy and sweet wrappers, residential litter, etc). Twelve times more non-agricultural pollutants were present in Szeged than in Szentes and twice as much as in Szarvas.

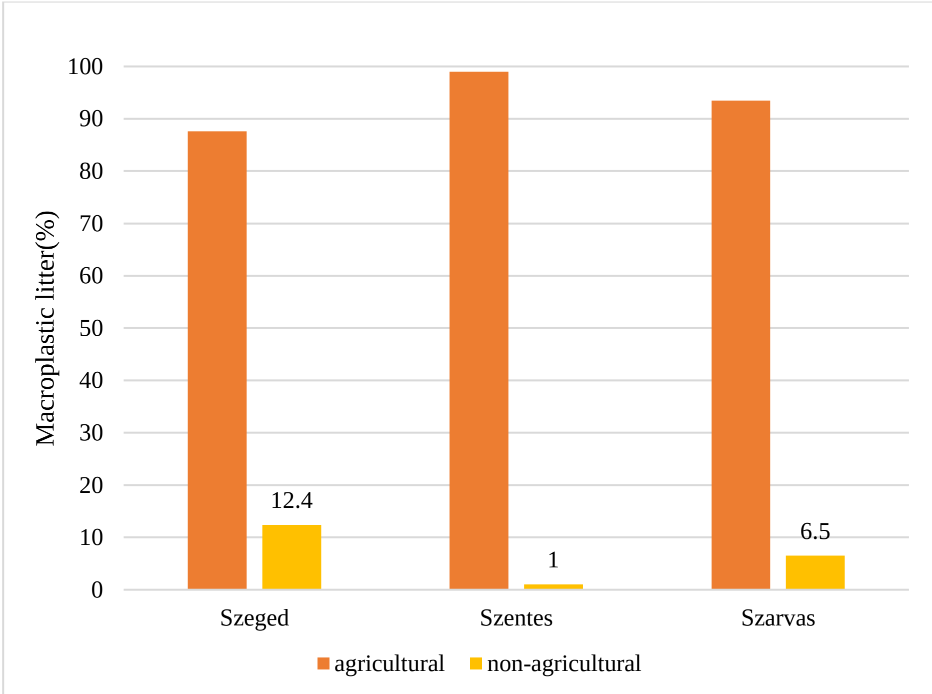

Figure 17. Abundance of macroplastics with agricultural and nonagricultural origin

Most agricultural products, including old films, broken pipes, and aged fibers that disintegrated as a result of weather, agrochemicals, and other variables, were directly connected to the production of macroplastics (Vox et al. 2008; Alhamdan and Alhilal, 2009; Babagyayou et al. 2020). Meanwhile, litter could be brought in from neighboring roads and rural areas by wind or runoff; thus, non-agricultural contaminants are present in the parcels due to unlawful trash deposition. Environmental elements including wind and run-off are critical in the movement and dispersal of macroplastics (Kundu et al. 2021). Wind transport was probably the most significant mobilizing force in our flat research sites. Szeged (12.4%) has the highest number of non-agricultural pollutants, followed by Szarvas (6.5%) and Szentes (1%). Because garbage along roadways is quickly transferred to nearby places by wind and water runoff, the Szeged and Szarvas greenhouses' proximity to major roads enhanced the high input of communal contaminants into the greenhouse areas, similar to the results of Choi et al (2020), Cao et al. (2021), and Stefano and Pleissner (2022). Last but not least, Szentes' low degree of pollutant contamination was due to its remote location and the long-term abandonment of its greenhouse farmlands; as a result, even workers probably did not litter there recently.

5.1.3. Morphological characteristics of macroplastics

a) Shape and Color

The macroplastics in the plots had the form of films, fragments, and fibers (Figure 13 A-C and figure18a). Plastic films made up the majority of the agricultural equipment; thus, films contributed to 74% –95% of the pollutants. Fragments were another significant plastic structure that made up about 15% –25% of the pollutants, but they were only found in Szeged and Szarvas. The least prevalent sort of contamination was fibers. Although it was noted at every location, it was not particularly common, as it made up only 1%–15% of the pollution. The areas with the highest and lowest fiber abundances respectively were Szentes, Szarvas, and Szeged.

A total of seven colors were recorded throughout the areas: transparent, gray, blue, white, black, red, and green (Figure 18b). There were six different colors documented in Szeged, three in Szentes, and five in Szarvas, indicating that the distribution of colors was not uniform. The most prevalent macroplastics (68%) were transparent, followed by those that were black (11%), gray (9%), white (6%), and blue (6%). Green and red were the two least frequently observed colors (2%).

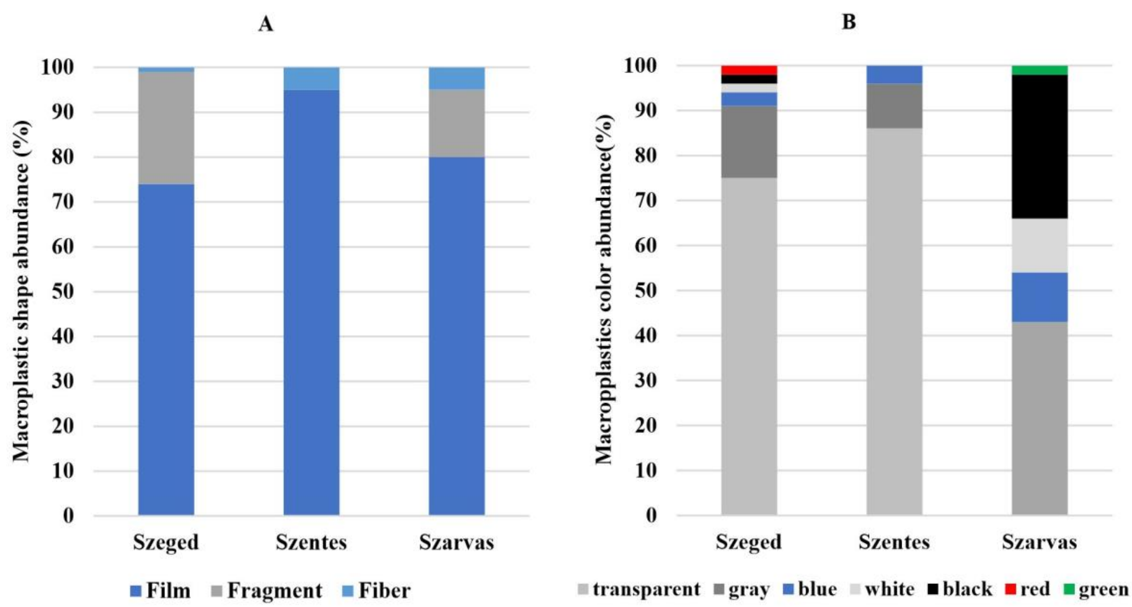

Figure 18. Morphological (a) and color types (b) of macroplastics

According to the findings, films were the most common shape of macroplastics, however, fibers and fragments were also present. The films inexorably came from the shattering of greenhouse cover materials, which is consistent with earlier research that identified plastic films as the primary microplastic structure in greenhouse soil (Kim et al. 2021; Yu et al. 2021; Liu et al. 2022; Wang et al. 2022). In mulching regions, plastic films were also the most prevalent pollutants (Liu et al. 2018; Meng et al. 2020; Huang et al. 2020). Due to the extensive usage of plastic films in greenhouse farming as well as the low durability and surface-clogging nature of plastic covers, they could be extremely abundant. The fact that non-agricultural contaminants, like food wrappers, can readily be transported by wind and water from major roadways and metropolitan areas to greenhouse farmlands is another factor contributing to the high quantity of films. Furthermore, these findings are consistent with the hypothesis that the majority of plastic pollutants in greenhouse farmlands arise from the fragmentation of old plastic coverings.

Contaminants in the area were found to be predominantly transparent, black, and gray in appearance. These colors match the colors of the plastic covers and PVC irrigation pipes that were used. The findings are consistent with earlier research that found various pollutant colors in agricultural soils. For instance, in Tanzania's and Germany's agricultural farmlands extensive colored pollutants were observed (Piehl et al. 2018; Kundu et al. 2021). The predominance of transparent materials (Figure 18b) may be attributable to greenhouses' demand for maximum light transmission for plant growth (Andrady, 2003); besides, they are also used because of their affordability, toughness, and lightweight (Patel and Tandel, 2017 and Sussana, 2018). Further, pieces of various colors were discovered on the ground, indicating that non-agricultural pollutants and other greenhouse plastic materials in the plots (e.g. boxes, compartments) were also fragmented and contributed to the production of macroplastic waste. Moreover, these color data help to clarify the connection between pollutants and greenhouse covers. Despite the fact that several colors were seen in the region, the color information reveals the sources of the contaminants and their connection to greenhouse films.

b) Polymer composition

Raman spectroscopy revealed the four main plastic types that could be seen on the plots were PE, PVC, PET, and PP (Figure 19b). PE, which made up 79%–93% of the overall abundance at all sites, was expected to be the most prevalent plastic contamination. On the other hand, PP was the least prevalent pollutant (2%–4%). All four polymers (PE, PVC, PET, and PP) were discovered in Szeged. When comparing their weights, PE was over four times more abundant than that of PVC, whereas the weight of PVC was six times higher than those of PET and PP altogether. Meanwhile, unlike in Szeged and Szarvas, only two polymers (PE and PP) were found in Szentes, where PE was nine times more abundant than PP. In Szarvas, PVC was three times more prevalent than PET and two times more prevalent than PP, while PE was nearly six times more prevalent and had the highest abundance.

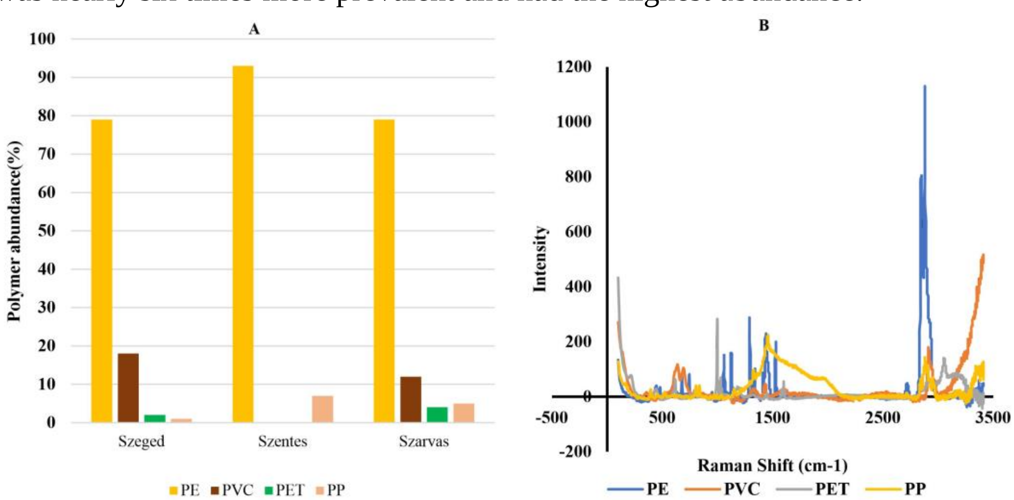

Figure 19. (A) Polymer compositions of the macroplastics collected on the plots of abandoned greenhouses. (B) Raman spectra of the identified polymer types.

These findings demonstrate that PE made up the most pollutants found in greenhouses. Because it is lightweight, malleable, and affordable, PE is the plastic material most frequently employed in greenhouse farming in the form of plastic films. Besides, it can help with disease and weed management (Andrady, 2003; Susanna, 2018; Liro et al. 2020). This result is consistent with earlier research (Liu et al. 2018; Meng et al. 2020; Huang et al. 2020; Wang et al 2022) that found PE to be the main pollutant on agricultural and horticultural surfaces. However, these results differ from those of Kundu et al. (2021) for the composition of plastic contaminants in the irrigated farmlands and riverbank of Arusha, Tanzania, where PET was the most abundant polymer. This discrepancy can be attributed, among other things, to the numerous contaminants that are introduced to riverbanks through litter, agricultural plastics, industrial waste, pharmaceuticals, and long-distance riverine transportation (Liro et al. 2020). In comparison, this research only focused on plastic pollutants with established sources and origins.

The quantity of macroplastics and the size of fragmentation are influenced by polymer composition. Due to PE's low density and ease of aging and fragmentation, the amount of microplastic particles on greenhouse agricultural surfaces subsequently grows. Meanwhile, as a result of their low abundance and minimal fragmentation, high-density polymers like PVC and PET also contribute fewer pollutants to greenhouse surfaces.

5.2. Microplastic abundance and characteristics

5.2.1 Abundance of microplastics in the soil surface and subsurface layers (0-40 cm)

Microplastics were found at most sampling points and soil depths in greenhouse farmlands. The overall total microplastic found in the area was 14100 pieces. The average microplastic abundance in the entire study areas was 440 pieces/kg.

The average microplastic contaminants abundance was 225 ± 61 pieces/kg in Szeged. The maximum abundance (500 pieces/kg) was recorded in four sites of parcels 1 and 2. While the least amount (0 pieces/kg) was recorded in parcels 2 and 3. Comparatively, the average microplastic contaminants abundance was 125±52 pieces/kg in Szentes. The highest contamination in the area was 400 pieces/kg found in the soil surface of a site of parcel 4 while the lowest abundance (0 pieces/kg) was recorded in the two sites of parcel 5. However, the result is much higher in Szarvas where an average abundance of 866± 102 pieces/kg was recorded in the entire area. The maximum abundance was 1500 pieces/kg recorded in a site of parcel 6 while the lowest abundance was 500 pieces/kg recorded in some sites of parcels 6,7, and 8 respectively.

The distribution of the contaminants varies with soil depth (Figure 20 and 21). In Szeged, the average microplastic contents in the surficial soil sample (0–20 cm) were 300 ± 93 pieces/kg, but in the deeper layer (20–40 cm) it decreased to 150 ± 76 pieces/kg. Microplastic contamination was higher in the surface layer of 3 sampling plots (parcels 1 and 2). However, only 1 sampling plot had greater microplastic abundance in the soil depth (Parcel 3). Equal distribution of microplastic contaminants was recorded in the 2 plots (in parcels 1 and 3). In Szentes, the number of microplastics decreased compared to Szeged, and in both surface and depths, it was the same (125 ± 94 and 125 ± 62 pieces/kg). Microplastic contaminants were only higher in 1 sampling plot (parcel 1). Two plots (2 and 4) show a higher abundance of microplastics in the sub-surface. Similarly, 2 points show equal distribution of microplastics in the soil depths. The most polluted soils were found in Szarvas, where in the upper soil layer the microplastic contamination was 1000 ± 163 pieces/kg, but in the deeper soil layer, it decreased to 733 ± 111 pieces/kg. Microplastic contamination was higher in the surface layers of four sampling plots (in parcels 6,7 and 8). Only 2 plots had higher microplastics in the sub-soil (plots 6 and 7). Thus, microplastic content was higher in the surface layer than in the deeper layer. However, the difference was not significant in all the areas (independent t-test: $P > 0.05$). However, the order of the microplastic abundance in the sites occurred in the following order: Szarvas greenhouse > Szeged greenhouse > Szentes greenhouse. Further, analysis of variance (ANOVA) revealed that microplastic abundance in Szarvas greenhouse is significantly different from Szeged and Szentes greenhouses ($P < 0.001$). On the other hand, no significant difference was reported between Szeged and Szentes ($P > 0.05$).

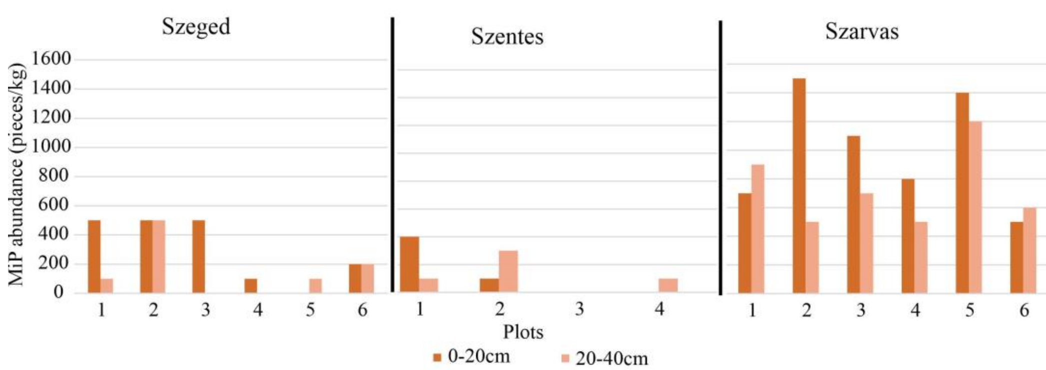

Figure 20: Abundance of microplastics in the sampling plots at 0-20 and 20-40 cm depths

Out of all the sampling points analyzed, the most contaminated one was found in Szarvas (1500 pieces/kg); while slightly contaminated sampling points were recorded in Szeged and Szentes (100 pieces/kg). There were three microplastic-free areas in the Szeged greenhouse, one sample was recorded from the surface layer (0-20 cm) while the other two were recorded at greater soil depths (20-40 cm). Similarly, three microplastic-free areas were recorded in Szentes, at two sampling points on the surface (0-20 cm) and one point in the deeper soil sample (20-40 cm).

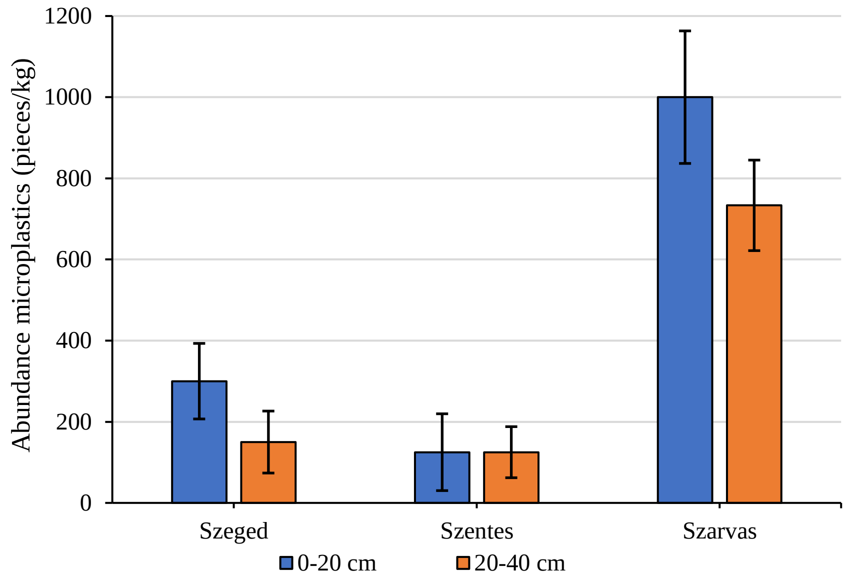

Figure 21: Microplastic abundance in the soil and sub-soil layer of the study areas.

The results on microplastic in the greenhouse soils agreed with the limited number of previous studies that found different concentrations of microplastic pollution in the soils of different regions under greenhouse farming. For instance, Isari et al. (2021) analyzed the greenhouse farmlands of tomatoes and watermelon in Ilia County, Western Greece, they found that the soils contained 301 and 69 pieces/kg respectively. The greenhouse soils in Yeoju, Republic of Korea, had an average of 755 pieces/kg (Choi et al., 2020). In contrast, Li et al. (2021) discovered 1,300–3,400 pieces/kg in Chinese greenhouse soils. Wang et al. (2022) also evaluated three different types of microplastics and came to the conclusion that abandoned greenhouses had the highest concentration of microplastics (2215.56 ±1549.86 pieces/kg), followed by regular (walled) greenhouses (891.11 ± 316.71 pieces/kg), and simple greenhouses (632.50 ± 566.93 pieces/kg). Liu et al. (2022) stated that the surface layers of agricultural lands contained 1813 ± 668 pieces/kg and the deep layers contained a similar amount (1875 ± 561 pieces/kg) of microplastic contaminations. Thus, the number of microplastics in the current study was lower than that in earlier studies. It could be explained by the non-application of waste sludge in the studied greenhouse farmlands, as Corradini et al. (2019) have shown that applying waste sludge increases the level of microplastic contaminants in the agricultural soil. The duration of greenhouse and mulch techniques, general differences across the study regions, the methods and management of land removal, and other factors could also be to blame for the discrepancy (Wang et al 2022).

However, when comparing the result of microplastics to macroplastics in the previous sub-chapter, we can see that the result corresponds. For example, Szarvas had the highest number of macroplastics, and as well as microplastics. The same goes for the other areas. Hence, we can conclude that macroplastic materials fragment into smaller pieces and pollute the soil with microplastic contaminants (Figure 22). This finding conforms to the previous studies which confirmed that the rate of litter fragmentation affects the availability of microplastics on the surface (Liu et al. 2018; Huang et al 2020). Besides the fragmentation, the other important sources of microplastic in the environment comprise surface water run-off, irrigation water, and atmospheric and wind deposition (Allen et al. 2019, Choi et al 2020; Rezaei et al. 2020; Katsumi et al. 2020; Wang et al. 2022).

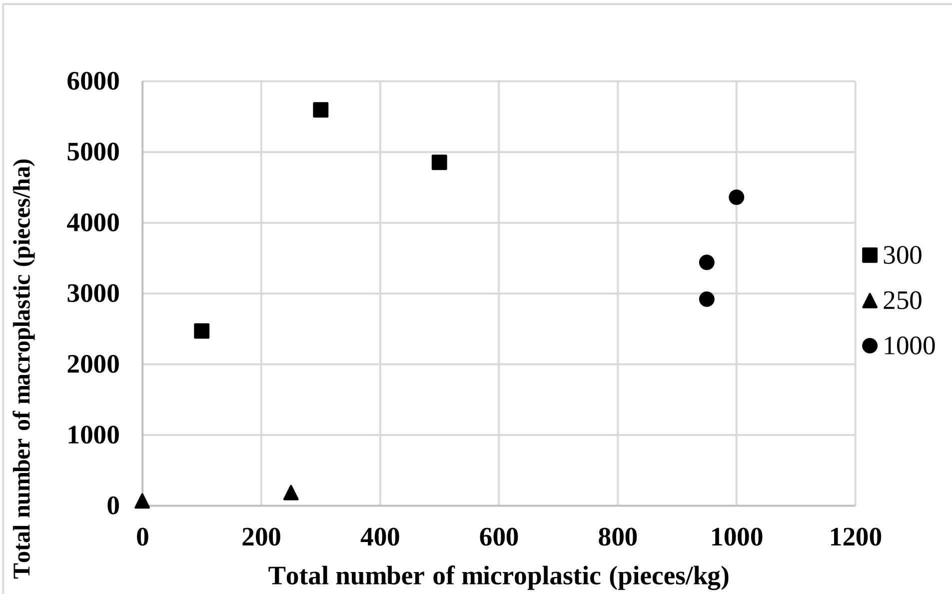

Figure 22: Macroplastic fragmentation to microplastic in the greenhouse environment

The reason for the high abundance of microplastic contaminants in the soil samples at Szarvas is because the area was recently abandoned (2017) compared with others and the fragmentation is ongoing as could be seen in the result of macroplastic in the previous chapter. On the other hand, the low abundance in the Szentes can be linked to the history of greenhouse abandonment which was longer compared to other areas. Similarly, the low abundance of microplastics in the area may be due to the area having shorter agricultural activities as they were mainly used for paprika seed production. This result of low abundance of plastic contamination in the old, abandoned greenhouse contradicts the previous findings of Wang et al. (2022) which concluded that the old, abandoned greenhouse had higher contents of microplastic contamination compared to other types of greenhouses. However, the variation may be due to the difference in greenhouse management, as Szentes study area was used for seed nurseries while the other farmlands were used for large commercial greenhouse farming.

Compared to the greenhouse sites, an average of 75 pieces/kg of microplastic contaminants were found in four control samples. The presence of microplastics in the control site, even though in small quantities, shows the ubiquitous nature and distribution of microplastics in all subsystems of the environment (Meixner et al. 2020). The low abundance of microplastic in the control soil samples reaffirmed that the microplastic contaminants found in the abandoned sites originated from the greenhouse plastic fragmentation as reported by previous studies that greenhouse farming heavily contributed to the microplastic contamination in soils (Isari et al. 2021, Yu et al. 2021; Fakour et al. 2021; Wang et al. 2022). However, the presence of microplastic contaminants in the control area may have occurred as a result of the atmospheric deposition, with the support of wind and precipitation (Rezaei et al. 2019; Allen et al. 2019; Haixin et al. 2022).

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

a) Size

The size of microplastic contaminants was divided into five categories (Figure 23A). Considering all sites, the most abundant size is 0-1 mm (49%) followed by the 1-2 mm size class (21%). The least size recorded was 4-5 mm (6%). In Szeged fallow, the most common size recorded was 2-3 mm (37%) followed by 1-2 mm size (25%). The least recorded size was 3-4 mm and 4-5 mm respectively. Contrarily, in Szentes fallow smaller microplastic items were more common (0-1 mm: 70%) followed by 1-2 mm (20%). The least size recorded in the area was 2-3 mm and 3-4 mm respectively. Similarly, the result of Szarvas also shows that 0-1 mm was the dominant size-class (67%), this is followed by 1-2 mm (17%), and the least size recorded in the area was 4-5 mm.

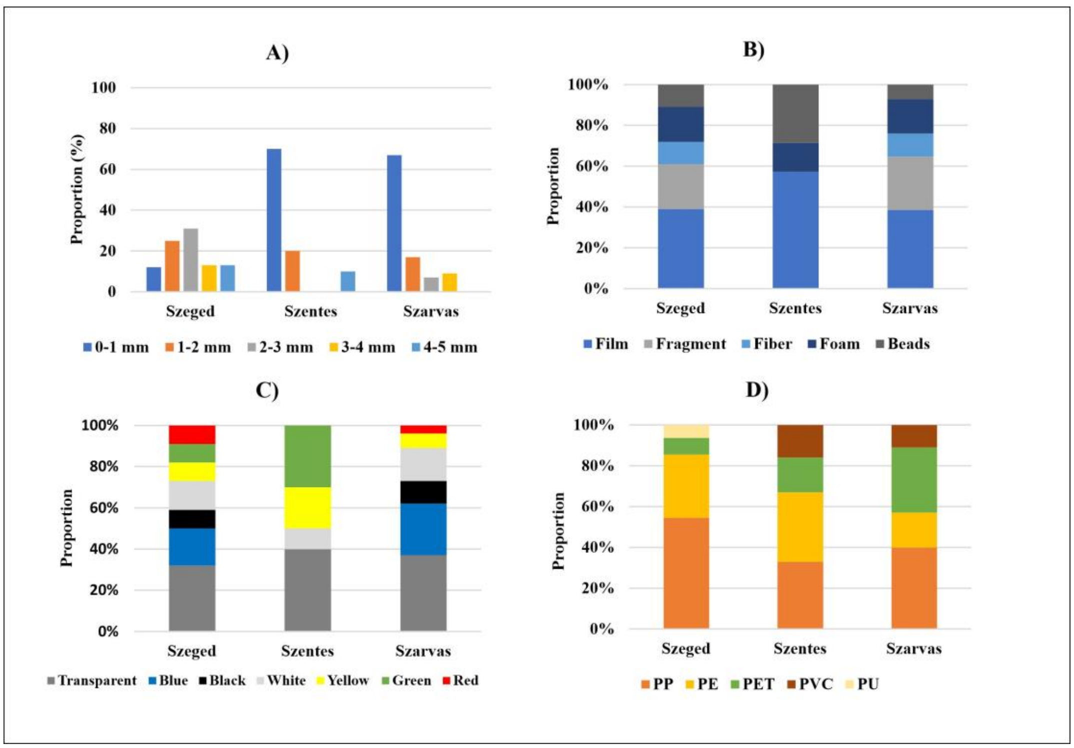

Figure 23: Main characteristics of microplastics

The different sizes of microplastic contaminants found in this study conform with the previous studies that recorded different categories of microplastic contaminants in mulching farmlands (Huang et al. 2020; Meng et al 2020). Similarly, previous studies about microplastic contamination in the greenhouse farmlands reported different categories of plastic contaminants (Choi et al 2020; Isari et al. 2021; Liu et al. 2022; Wang et al. 2022). The microplastic contaminants classes found in this study area and other greenhouse environments could be explained by the various degrees of fragmentation of the larger plastic materials in the greenhouse environment (Berenstein et al. 2024). The fragmentation could be triggered by climatic factors, environmental contaminants agrochemical usage, and the structure of the greenhouse environment (Dilara and Briassoulis, 2000; Vox et al. 2008; Dehbi et al. 2015). Furthermore, the size of microplastic found in the areas may be related to the local factors in the study areas such as climatic factors, level of pollutants, aging, and agrochemicals usage. All these factors contribute to the creation of plastic fragments of various sizes. Different studies confirmed that the larger macroplastics and mesoplastics in the greenhouse environment and mulching do fragment into smaller pieces and end up in the soil ecosystem (Li et al. 2020; Haixin et al. 2022; Wang et al. 2022; Berenstein et al. 2024). However, the distribution of microplastic size is not uniform in the soil horizon as larger particles concentrate on the soil surface; the smaller ones migrate easily to the deeper soil zones (Yu et al. 2021). Likewise, in this study, the bigger size microplastic contaminants were recovered in the soil surface (0-20 cm), while the smaller microplastic contaminants were recovered in the soil sub-surface (20-40 cm). The reason for the distribution disparity in the soil depths may be due to the surface layers directly receiving fragmented plastic contaminants, and only a few smaller microplastics get into the soil because of land management, soil texture, and cracks in the soil.

b) Shape

Five different shapes of microplastics were identified (Figure 23B). Considering all study areas, microplastic fibers dominated (44%), followed by films (29%), fragments (18%), beads (7%), and foam (2%). Only three morpho-types were recorded in the Szeged greenhouse area: the average percentage of fiber was higher (60%), followed by microplastic films (34%), and the least recorded shape in the area was foam (6%). Similarly, three shapes were recovered in Szentes. The average percentage shows uniformity, as both fiber, film, and fragment were equally distributed (33% each). However, four shapes were recorded in Szarvas, where fiber was the most common shape (40%) followed by fragments (22%), beads (21%), and films (17%).

The results on the shape of the microplastic show that greenhouse soils are characterized by different contaminant shapes, of which microplastic fibers, films, and fragments are dominant morpho-types found in the abandoned greenhouse environment. This result is consistent with the previous studies that found different shapes (such as fibers, films, fragments, beads, and foam) of microplastics in the greenhouse environment (Li et al. 2019; Yu et al. 2021; Kim et al. 2021; Wang et al. 2022; Liu et al. 2022). According to previous studies, the major contaminant structure found in the greenhouse environment were fiber, fragments, and films (Zhang et al. 2018; Chen et al. 2020; Ding et al 2020; Kim et al. 2021). Similar results were found in this study, the high abundance of microplastic fiber, film, and fragments in the soils might be associated with the weathering of larger plastics that were used as primary materials in greenhouse farming, so the fibers originate from the macroplastic fiber threads used for tightening greenhouse structure. Films from plastic foil are used for greenhouse coverage and fragments from the broken pipes, crates and other plastic materials. This was confirmed in previous studies which reported that larger macroplastic materials weathered because of several factors and form soil microplastic (Huang et al. 2020; Wang et al 2022; Liro et al. 2023; Berenstein et al. 2024). Similarly, the previous chapter on macroplastic confirms this claim by revealing how macroplastic weathers into pieces and ends up in the soil environment. However, due to the long abandonment period of these farmlands, atmospheric deposition, water runoff, and wind might be another important source of microplastics, especially fibers (Rezaei et al. 2019; Allen et al. 2019; Haixin et al. 2022).

c) Color

Microplastics appear in a wide range of colors (Figure 23C), comprising white, yellow, blue, black, transparent, green, and red. The overall color distribution in the area shows that according to the study, in all areas transparent color was the dominant color (35%), followed by blue (15%), white and green colors (13-13%), yellow, (12%), black (7%) and red (4%). The result of Szeged shows that transparent was the dominant color (32%), followed by blue (18%), white (14%), black, yellow, green, and red (9% each). On the other hand, only four colors were recorded in Szentes, of which the dominant one was transparent (40%) followed by green (30%), yellow (20%), and white (10%). However, highly diverse colors were recorded in the soil of Szarvas greenhouse. Here, transparent color was the dominant (37%), followed by blue (26%), and white (16%). Others include black (11%), and yellow (7%). The least color found was red (4%).

The result of microplastic color is very important as it reveals the nature and sources of microplastic contamination. In this study, the result shows that the microplastic contaminants were highly diverse in terms of color. This result tallies with the previous findings which have shown that microplastic contaminants in the agricultural soils were made up of different colors (Liu et al. 2018; Yang et al. 2021). For example, a wide range of colors was discovered in the conventional agricultural soils of Germany (Piehl et al. 2018). Similarly, Meng et al. (2020) and Huang et al. (2020) discovered different colors in the mulch soils of China. Isari et al. (2021) reported that microplastic contaminants in greenhouse soils relate to the plastic used as coverage. Contrarily, Boughattas et al. (2021) and Wang et al. (2022) reported over 7 different colors in the soils of a greenhouse environment. Microplastic in the greenhouse environment can be the product of the larger macroplastic used for covering, as these macroplastic items weather driven by climatic factors, agrochemical usage, and environmental pollution (Andrady, 2000, Vox et al. 2008; Babagyayou et al. 2020).

However, the various colors can originate from external sources as well, due to atmospheric deposition, wind action, and surface runoff (Choi et al. 2020; Liro et al. 2022).

The dominant transparent color in this study shows the link between the plastic films used for coverage and microplastics. For example, 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. Other colors found in the areas show that external contamination may occur in the areas, especially at Szeged and Szarvas.

d) Polymer composition

A total of 5 different polymer types were recorded in this study: PE, PP, PU, PVC, and PET (Figure 23D). The overall frequency of the polymer types reflects that PP was the dominant polymer (44%), followed by PE (29%), PET (16%), PVC (9%), and PU (2%). In Szeged only three polymer chemical compositions were recorded: PP (60%), PE (34%), and PU (6%). On the other hand, four polymer compositions were recorded in Szentes greenhouse: PE (34%), PP (33%), PET (17%), and PVC (16%). Meanwhile, three microplastic polymer compositions were recorded in Szarvas greenhouse: PP (40%), PET (32%), and PVC (11%).

Previous studies confirmed the presence of different polymer types in agricultural soils. Liu et al. (2018) confirmed the presence of nine polymer types in the agricultural soils of China. These polymers were PP, PE, PA, PET, PVC, PC, Acrylonitrile butadiene styrene, PMMA, and PS. Ding et al. (2020), recorded. PS, PE, PP, HDPE, PVC, and PET in the conventional farmlands. Piehl et al (2018) found 7 polymer types in the agricultural soil that were not amended with organic fertilizers in Germany. The recorded polymer types were PS, PET, PMMA, PVC, PP, and PE. However, Wang et al. (2022) recorded over 10 polymer types in the greenhouse soils: PP, PE, Polypropylene polyethylene copolymer (PP: PE), Poly octadecyl acrylate, Poly 1-tetradecane, PET, Polyacrylonitrile: acrylic acid, PS, and Rayon. However, in this study, PE and PP were the most identified polymer types. This conforms with previous findings which showed that despite the numerous polymer types found in the greenhouse farmlands, PE and PP were the most frequent polymer identified (Liu et al 2018; Boughattas et al. 2021; Kim et al. 2021; Yu et al. 2021; Wang et al. 2022).

Furthermore, this study differs from other studies in terms of the number of identified polymer types. Some studies (such as Liu et al. 2018; Boughattas et al. 2021; Kim et al. 2021) recorded several polymer types, and the high number of recorded polymer types in these areas compared to this study occurred as a result of contamination by external sources. For example, the high number of polymer types recorded in the suburbs of China by Liu et al. (2018) might be associated with the proximity of these lands to urban regions and external contamination because of anthropogenic factors.

The reason for the high abundance of PE in these study areas was that this polymer type was mainly used as a greenhouse film for greenhouse structure coverage. In addition, PP was highly recovered in the areas because plastic fibers that were made up of PP chemical composition were also used in the areas as threads to hold on greenhouse structures. Besides, PP fibers were used for tightening the containers of the final agricultural products. Other polymer materials such as PET and PVC have occurred in the areas as a result of the broken water pipelines and crates for packing and transporting vegetable products. However, apart from the above-mentioned agricultural activities, another source of polymer composition can be external as the previous chapter showed that 10% of the total macroplastic comes from external sources and they also could weather in the soil.

5.3. Abundance and distribution of microplastic in the entire profile

Microplastic is extensively distributed in the layers of the profiles (Figure 24). The mean abundance of microplastic was compared by One-way ANOVA and the result showed that there were significant differences in the microplastic contents in the soil profiles of the areas [$F(2, 66) = 4.59$, $P= 0.014$]. The average microplastic abundance varied among the profiles analyzed were 80 pieces/kg, 4 pieces/kg, and 96 pieces/kg, for Szeged, Szentes, and Szarvas respectively. It can be revealed that microplastic abundance was higher in Szeged and Szarva's profiles than in Szente's profiles. The differences were found to be highly significant among them (p values < 0.05).

The result of grain size distribution showed that the soil particle sizes and arrangements of the three areas are similar with little difference (Figure 24). Medium coarse silt, clay, and coarse silt were the dominant textures. Also, the medium (15.6-31 µm) and coarse silt (31-63 µm) were more dominant in the entire soils of the profiles. In Szeged, individual profile analysis revealed that the distribution of microplastic particles was not uniform; hence Profile 1 had the highest concentration (200 pieces/kg) of microplastic in depth 100-120 cm. While the 100 pieces/kg were respectively recorded in 40-60 cm, 120-140 cm, and 140-160 cm. According to Spearman's correlation analysis, there was a moderate positive correlation between depth and microplastic content in this profile, but it was not statistically significant [$r(8) = 0.626$, $P = 0.097$]. The medium (15.6-31 µm) and coarse silt (31-63 µm) particles predominantly cover the profile and account for about 64%. On the other hand, profile 2 has the highest microplastic concentration (80-100 cm: 300 pieces/kg) followed by 0-20 cm with a microplastic concentration of 200 pieces/kg. There was a weak negative correlation, which was not statistically significant, between depth and microplastic content [Spearman's correlation: $r(6) = -0.235$, $P = 0.653$]. The medium (15.6-31 µm) and coarse silt (31-63 µm) particles accounted for about 70% of the total grain-size structure. Further, in profile 3, the highest concentration of 300 pieces/kg was recorded in a 40-60 cm layer followed by 0-20 cm with 200 pieces/kg. There was a strong negative correlation between depth and microplastic content, which was not statistically significant [$r(6) = -0.759$, $P = 0.080$]. The grain size distribution was predominantly the medium (15.6-31 µm) and coarse silt (31-63 µm) accounting for over 64%.

In Szentes, two individual profiles were also analyzed. In Profile 1, the uppermost layer was found to be contaminated (100 pieces/kg) while the other layers beneath were free from microplastic contamination. Thus, according to Spearman's correlation analysis, there was a moderate negative correlation between depth and microplastic content in this profile, but it was not statistically significant [r (11) =-0.500, P =0.117]. Also, the medium (15.6-31 µm) and coarse silt (31-63 µm) accounted for 43% of the total grain size distribution while the clay and very fine silt particles accounted for about 22% and 10% respectively. The surface layers had high contents of clay compared to the deeper layers. On the other hand, in profile 2 the medium (15.6-31 µm) and coarse silt (31-63 µm) accounted for 40% of the total grain size particles. The clay and very fine silt particles accounted for 24% and 12% respectively. High content of clay and silt were recorded on the first layers of the soil profile. The level of the clay and very fine silt particles decrease as depths increase. Hence there was no abundance of microplastic in all the layers of the profile.

Similarly, in Szarvas, the result of microplastic distribution in Profile 1 shows that the highest microplastic concentration (500 pieces/kg) was recorded in 20-40 cm, followed by 0-20 cm with 300 pieces/kg. The Spearman correlation analysis shows that there was a moderate negative correlation between depth and microplastic content in this profile, but it was not statistically significant [r (4) =-0.316, P =0.684]. The medium (15.6-31 µm) and coarse silt (31-63 µm) particles accounted for 65% of the total grain size distribution in the profile. However, the clay and very fine silt particles accounted for 25% of the total particles in the profile. The clay content increased with depth; thus, the surface layers had a low level of clay content compared to the deep layers. Likewise, Profile 2 had the highest microplastic concentration (300 pieces/kg) in the 0-20cm layer followed by 200 pieces/kg in 20-40 cm. According to Spearman's correlation analysis, there was a moderate negative correlation between depth and microplastic content in this profile, The relationship was not statistically significant [r(10) =-0.555, P =0.096]. The medium (15.6-31 µm) and coarse silt (31-63 µm) particles accounted for 65%. The clay and very fine silt particles accounted for 34%. These particles were recorded lower on the surface than on profile depth. Similarly, Profile 3 had the highest concentration of 400 pieces/kg in 0-20 and 40-60 cm layers respectively, followed by 100 pieces/kg in 80-100 cm layers. According to Spearman's correlation analysis, there was a strong negative correlation between depth and microplastic content in this profile [r (13) =-0.697, P =0.008]. The relationship is statistically significant. However, the medium (15.6-31 µm) and coarse silt (31-63 µm) particles accounted for about 62% while the clay and very fine silt particles accounted for 37%. Like the other profiles in the area, the clay and fine silt particles were higher on the depths than on the profile surface.

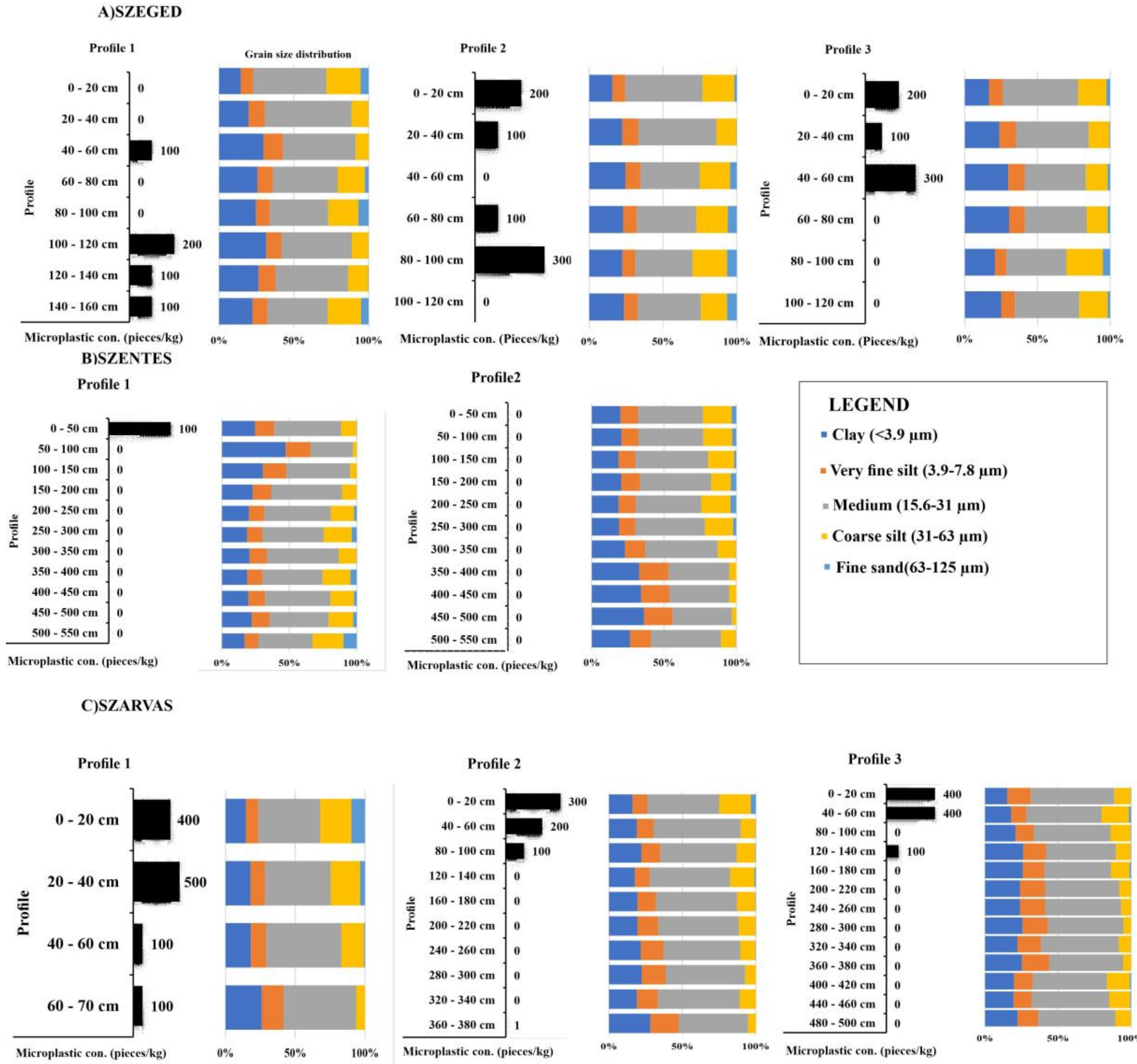

Figure 24: Abundance and distribution of microplastic in the soil profiles

The result of microplastic distribution in the soil profiles shows that microplastic pollutes different layers of soil profiles. This result supports the previous studies that confirmed the pollution of microplastic in the different depths of soil profiles (Zhang et al. 2019; Lu et al. 2021; Meng et al. 2022; Tagg et al. 2022). Li et al. (2022) reported that microplastic pollution occurred in 20 cm depths of agricultural areas. Hidayaturrahman et al. (2019) and Harms et al. (2021) concluded that microplastic contamination occurred down to 30 cm layers of agricultural areas. Further, Cao et al. (2021) studied the vertical distribution of microplastic in the soils of vegetable gardens, paddy, and maize fields. Their result showed that microplastic particles were recorded down to 80 cm depths of the soils. The result of my study is different from the previous studies as I confirmed the microplastic contamination in the greater soil depths (down to 160 cm) against the findings of the previous studies which only limited to 80 cm contamination.

The distribution of microplastic in the soil depths was not uniform in the study areas. These differences can be linked to the grain size distribution of the soil in the study area (Figure 24). For instance, at Szentes the surface layers have high contents of clay (<3.9 µm) and very fine silt (3.9-7.8 µm) which largely impede the vertical movement of contaminants through them (Yu et al. 2021). Contrarily, the high abundance of microplastic in the layers of Szeged and Szarvas profiles can be a result of the nature of the particle size distribution of the profiles as the surface layers contain less clay and very fine silt particles, hence the rate of plastic migration is presumably to be higher in the pores of larger grain size particles (O'Connor et al. 2019; Schell et al. 2022). Apart from the soil texture and particle size distribution, the differences in the microplastic abundance in the soil depths can also be traced to the differences in agricultural practices (Meng et al. 2020; Schothorst et al. 2021). The greenhouse farmland at Szentes was subjected to light greenhouse cultivation because the area was used for a pepper nursery site while the other areas were subjected to full greenhouse farming in large quantities. Another factor that influences this difference is the duration of greenhouse farming, which might serve as the reason for these differences (Li et al. 2020; Kumar and Sheela 2021; Zhang et al. 2022). For example, Szentes's greenhouse operated only for 21 years, while Szeged and Szarvas operated for 25 and 27 years respectively.

Furthermore, the presence of cracks on the soil surface is another factor that may favor microplastic migration in the soil depths. Cracks were reported through which contaminants such as microplastics penetrate the profile layers (Zhao et al 2022). Because of the high content of clay minerals on the surface of the soils as revealed by grain size analysis, also several cracks were observed during the sampling period of this study, and these cracks may probably serve as pathways that microplastic penetrated and polluted the deeper zones of the soil profiles of these areas. Also, the nature of agricultural activities in the area is another primary factor that favors microplastic contamination in the agricultural soil (Meng et al. 2020; Harms et al. 2021). Agricultural practices such as harrowing, plowing, and ridges expose the soil depths and bury the soil surface providing potential carriers of microplastics. Besides, leaching, the effect of plants and animals through burrowing and plant entanglements affect the vertical migration of microplastics in the soil depths (Bläsing and Amelung, 2018; Li et al. 2022)

5.3.1. Morphological characteristics of microplastic contaminants of the entire profiles

As different microplastic shapes (film, fiber, fragment, bead, foam,) are found on the soil surface, four different shapes of microplastics were identified in the soil profiles of Szeged and Szarvas (Figure 25). These are fiber, film, fragment, and bead. Considering all the profiles, microplastic fiber dominated the area (72%), followed by fragments (19%), beads (6%), and films (3%). The morphological structure of Szente's contaminants was not included in this analysis because only one yellow fragment particle was recorded in the entire profile. However, the microplastics found in the profiles appear in a wide range of colors. The overall color distribution in the two areas showed transparent color as the dominant color (37%), followed by red (23%), blue (20%), white (14%), and black (6%). Importantly, all the microplastics found in the soil profiles were very small in size. Apart from their counting, no other analyses were carried out.

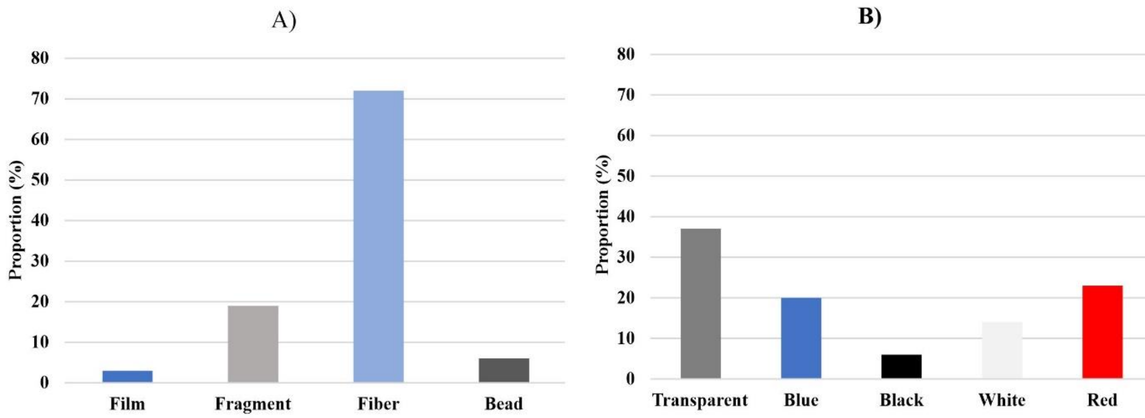

Figure 25: Main characteristics of microplastics in the entire profiles

The result of different morphological structures found in the contaminants of the entire soil profiles conforms with the previous findings that discovered different shapes in the soil depths. Different scholars explored the soil and soil profiles of agricultural areas in different parts of the world and suggested that different morpho-types of microplastics were found in the soil depths, these shape structures include fiber, film, fragment, beads, film, pellet, and foam (Cao et al. 2021; Fakour et al 2021; Tagg et al. 2022). The high abundance of fiber in this study corresponds with previous studies that reported that fibers were the major contaminant type found in the soil depth (Liu et al. 2022; Yu et al. 2022). The reason for the high abundance of fiber in the soil depths is that fibrous materials are one of the major contaminant structures found in the greenhouse environment (Zhang et al. 2018; Chen et al. 2020; Ding et al 2020; Kim et al. 2021). On top of that, fibrous contaminants easily penetrate the soil pores and cracks (Cao et al. 2021; Ragoobur et al. 2021). Further, because of their light nature, they can be easily carried out and transported by water as it serves as the main agent for transporting microplastics to the soil depths (O'Connor et al. 2019; Mora et al. 2021). On the other hand, fragments were the second type found in the soil depths of this study. This conforms to the findings of Cao et al. (2021) and Fakour et al. (2021) which reported that fragment is the main structure found in soil depths. Fragment structure occurred due to the weathering of larger macroplastic materials (Liro et al. 2023; Berenstein et al. 2024). They migrate to the soil depth through large pore sizes and cracks in the soil surface.

Moreover, the result shows that the microplastic contaminants in the soil depths are composed of highly diverse colors. This result tallies with the previous findings which have shown that microplastic contaminants in the soil surface and soil profiles made up of different colors (Cao et al. 2021; Tagg et al. 2022). The result of color shows the relationship between macroplastic and microplastic contaminations, it also confirms that microplastic contaminants from the soil surface do migrate to the soil depth. However, compared to the surface layers, the size of microplastics found in the soil depths is very small. This conforms with the previous studies which reported that most of the microplastic contaminants found in the soil depth appear to be smaller in size compared to what is obtainable on the soil surface (Tagg et al. 2021; Yu et al. 2021). The reason for the presence of smaller microplastic contaminants in the soil is that larger microplastic materials find it difficult to penetrate the soil and reach the soil depths.

5.4. Soil physical parameters and microplastic distribution of the entire profile

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

Particle size distribution of the soil is a very important parameter in determining the distribution and migration of pollutants in the soil. Considering all collected soil samples, the overall result of the grain size distribution shows that the particle size compositions of the soils in all three study areas are similar, as they have coarse silt textures. Further categorization of the soils reveals that the soils are predominantly silt loam and silty clay loam. Correlation analysis between microplastic and clay showed a weak negative correlation but was statistically significant (Table 3). Similarly, the result shows a significant weak negative correlation between microplastic abundance and very fine silt particles. Contrarily, a significant weak positive correlation was recorded between microplastic abundance and medium silt particles. The relationship between microplastic abundance and coarse silt was a very weak positive correlation. Even though, the relationship was not statistically significant. Further, the result revealed that the relationship between microplastic abundance and fine sand particles is a very weak negative correlation, the relationship was not statistically significant.

Table 3: Correlation between microplastic and soil textural classes

S/n	Variables	Microplastic
1	Microplastic	-
2	Clay	0.28^{**}
3	Very fine silt	0.23^{*}
4	Medium silt	0.25^{*}
5	Coarse silt	0.15
6	Fine sand	0.12

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

Four textural soil classes were identified in the study areas: silt loam, silty clay loam, silty clay, and clay loam. Among these textural classes, only silt loam and silty clay loam had microplastic abundance (Figure 26). Silt loam had an average of 200± 48 pieces/kg 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. Importantly, the contaminant distributions were found to be higher in the upper soil layers and less abundant in the soil depths. This is well portrayed in Szentes and Szarvas farmlands.

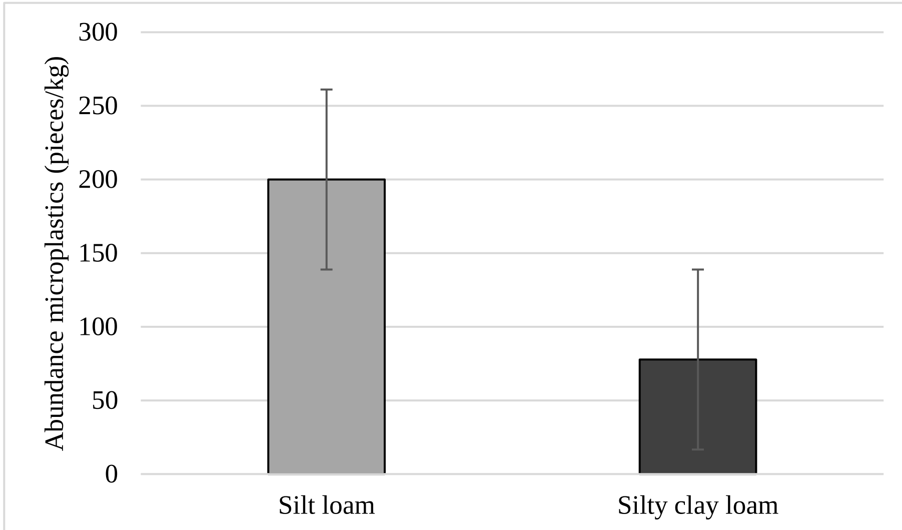

Figure 26: Microplastic abundance in the soil textural classes

The classification of soil particle size distribution shows the soils of the entire study areas were mainly similar. The silt particles (very fine, medium, and coarse: 3.9-63 µm) dominated the samples and accounted for about 75% of the grains, while the clay particles accounted for 20% and sand materials accounted for about 5%. Generally, soil particles can be classified as coarse silt. This result of the distribution of microplastics in the silt loam textural class conforms with the previous finding of Yu et al. (2021) which compared the distributions of microplastics in the different soil textural classes and concluded that the distribution was not uniform as loam and silt loam textural classes dominated the distribution.

Moreover, the correlation result of particle size distribution shows that the grain-size particle distribution of the studied soils slightly favors the easy migration of microplastics. This result contradicts the previous findings which reported that microplastic positively correlated with soil textural classes and that microplastics easily migrate vertically in the soil profiles and sediments (O'Connor et al. 2019; Lu et al. 2021; Cao et al. 2021; Ding et al. 2021; Zhang et al. 2022). The reason for the variation of this finding with previous studies is that the soils analyzed in previous studies were predominantly sandy in textures. Sandy and coarser soils permit the movement of microplastics through them (Watteau et al. 2018; Yu et al. 2021). For example, over 82% of the soil samples analyzed by Zhang et al. (2022) were sandy with grain sizes of 516-754 µm, and their clay and silt content accounted only for 0.7% and 17% respectively. Similarly, larger sand particles (516 and 754 µm) dominated the soil texture in the study of O'Connor et al. (2019). Therefore, the presence of microplastics in some parts of the studied profiles partly occurred as a result coarseness of the soil and the presence of cracks in the soil as it was widely observed in the areas during the fieldwork. In support of this, Zhao et al (2022) stated that cracks in the soil serve as a pathway for microplastic migration into the soil profile. Furthermore, the long abandonment of the areas as well as farm management practices may also serve as reasons for profile contamination.

5.4.2. Porosity, bulk density, saturated hydraulic conductivity of the soils and microplastic distribution

Soil physical parameters may affect the distribution of microplastics at various depths in the soil profiles (Table 4). Out of 13 soil samples analyzed for porosity, 9 samples show normal compaction levels (40-50%), and only 4 samples show a high level of compaction (< 40%). Hence, the soils of the study areas can be categorized as normal, and the movement of water and other contaminants especially through the surface layers is possible. Most of the non-compacted layers were recorded on the surface layers, while the compacted layers were recorded in the deeper soil depths (50-60 cm, 70-80 cm, 80-90 cm, and 100-110 cm).

In addition, the bulk density result shows that most of the analyzed soils (9 out of 13) fell in the non-compacted category, while only a few samples (3 samples) were compacted. The non-compacted layers were recorded in the surface layers while the compacted ones were recorded in the deeper layers (50-60 cm, 70-80 cm, and 100-110 cm). The restriction of downward movement of microplastic can be seen as the vertical migration of microplastics heavily reduced from 133 pieces/kg to 33 pieces/kg in the compacted layer (Szeged: 70-80 cm) simultaneously with porosity and bulk density changes.

Table 4: Soil physical parameters

S/n	Location	Depth (cm)	Porosity (%)	Bulk density (g/cm^3)	Saturated hydraulic conductivity (m/s)	Average microplastic abundance (pieces/kg)
1	Szeged	10-20	40.86	1.59	9.4×10^{-10}	133
2	Szeged	30-40	40.50	1.52	4.0×10^{-7}	66
3	Szeged	50-60	35.41	1.69	9.4×10^{-10}	133
4	Szeged	70-80	36.68	1.75	2.8×10^{-9}	33
5	Szentes	10-20	42.86	1.53	1.98×10^{-9}	50
6	Szentes	30-40	42.56	1.55	9.4×10^{-10}	50
7	Szentes	50-60	44.16	1.53	2.0×10^{-9}	50
8	Szarvas	10-20	46.77	1.30	1.5×10^{-9}	366
9	Szarvas	30-40	42.25	1.47	1.5×10^{-9}	N/A
10	Szarvas	50-60	42.74	1.52	1.5×10^{-9}	233
11	Szarvas	70-80	40.42	1.56	1.5×10^{-9}	N/A
12	Szarvas	80-90	38.97	1.59	1.5×10^{-9}	50
13	Szarvas	100-110	38.40	1.62	1.5×10^{-9}	N/A

However, the result of saturated hydraulic conductivity shows that most samples analyzed are impermeable, and aquitard. Out of 13 samples, only one sub-surface layer (Szeged: 30-40 cm) was found to be permeable. The non-aquitard layer retained very few microplastic contaminants (66 pieces/kg) and let pass plentiful microplastic contaminants (133 pieces/kg). To conclude, we can say that the result of the soil's physical parameters proves that microplastic penetrates the soil layers and passes through the soil pores with the aid of water movement as reported by previous studies (O'Connor et al. 2019; Oßmann et al. 2021; Schell et al. 2022).

The results of soil porosity and bulk density show that the soils in this study area permit the movement of microplastic especially in the surface layers. In Szeged, the surface layers (0-40 cm) have normal porosity and bulk density that allow the movement of water. Presumably, microplastics could migrate downward through these pores to the soil depths. However, the beneath layers stop the movement of water and microplastic because the rate of penetration is very little or not possible due to the level of compaction. However, in Szentes, the porosity and bulk density result was uniform, hence the movement of water and other contaminants tends to be uniform throughout the depths of the soil. At Szarvas the result was found to be similar to Szeged's as the surface layers (10-80 cm) were recorded to be normal in porosity and bulk density while the subsurface layer (80-110) was recorded to be compacted. This result of microplastic migration in the surface layers due to high porosity and bulk density conforms with the previous finding that reported the vertical migration of microplastics in the soil profiles, sediments, and groundwater (Oßmann, 2021; O’Connor et al., 2019; Schell et al., 2022). Contrarily, the soil's low hydraulic conductivity impeded the vertical migration of water and contaminants in the soil except in the non-aquitard layer (30-40 cm) in Szeged where the soil retains very few microplastic contaminants (66 pieces/kg) and let passes plentiful microplastic contaminants (133 pieces/kg). However, since the study reveals that the soil's physical properties did not support the favorable vertical migration of the microplastic contaminants, probably, other processes such as infiltration help the water and microplastic to migrate through the soil pores (O'Connor et al. 2019; Oßmann et al. 2021; Schell et al. 2022).

6. CONCLUSION

Plastic contamination of the environmental systems is one of the hot topics that intrigues researchers in the 21st century, and there is a need to carry out studies to reveal the nature of the problem and provide methodological solutions to this problem. The result of this study reveals the actual level of macroplastic and microplastic contamination in the greenhouse environment. This study is among the few that attempted to look at the available methodologies and suggested an economical and effective method for extracting microplastic from soil with a high content of organic matter. This study, however, quantifies the level of microplastic contamination in the greenhouse environment. Further, the study filled in the gap on how macroplastic materials weather to become microplastic particles, as well as how these particles pollute the vertical profile of greenhouse soils. Also, it is among the few studies that revealed how the soil structures and other properties influence the vertical movement of microplastics in the soil depths.

Again, the findings of this study confirmed the existing knowledge about microplastic contamination in the agricultural environment. It also confirmed the hypothesis of previous findings about the contribution of greenhouse farming in the production of plastic contaminants to the environment. Nevertheless, the findings may have practical applications in greenhouse farming cleaning, monitoring, and plastic reuse and recycling activities.

This chapter concludes the research findings considering the research objectives. Further, the chapter also briefly explains the contribution of this study to the field of plastic contamination and monitoring. The chapter also reflects some limitations and offers possible recommendations for future studies in the fields of macroplastic and microplastic contamination in agricultural areas.

6.1 Macroplastic litter in fallow greenhouse farmlands

The study aimed to quantify the level of macroplastic in the abandoned greenhouse farmlands. The results indicate that macroplastic litter was discovered in all sampled parcels and contaminated the abandoned greenhouse farmlands with an overall mean abundance of 2655 pieces/ha. The highest number of pieces (mean: 4328 pieces/ha) was found in Szeged greenhouse farmlands; thus, the greenhouses were polluted by 5 kg/ha macroplastic litter. The plastic contamination had a greater weight (6.4 kg/ha) in the greenhouses at Szarvas, but as the pieces were larger, the mean number of macroplastic pollution (3513 pieces/h) was lower compared to Szeged. Contrarily, the lowest number of pieces was low at Szentes (150 pieces/ha). The level of plastic contamination was 34 and 28 times lower than Szeged and Szarvas.

The level of contamination in the study areas was higher than what was reported in the agricultural farmlands of other countries such as some parts of Germany and Switzerland (Pehl et al. 2018; Kawecki and Nowack, 2021). On the other hand, the level of contamination was lower than what was reported in China (Huang et al. 2020) and in other parts of Germany (Stefano and Pleissner, 2022). So, the contamination could be considered as a medium in the study areas.

The results show that macroplastic litter weathers to microplastic through fragmentation. The process of fragmentation is still ongoing, especially in Szarvas because there are larger macroplastic particles that were not broken, probably they will contribute to the next fragmentation. Thus, the degree of fragmentation is small at the Szarvas greenhouse, followed by the Szeged greenhouse, and finally Szentes greenhouse. These differences occur because the amount of temperature, and solar radiation were lower in Szarvas than in Szeged and Szentes.

This study was among the first set of studies that revealed that only 90% of contaminants found in the greenhouse farmlands had agricultural origin. These contaminants originated from the fragmentation of plastic film for coverage, fiber-tightening structures, and fragments for water pipes. Similarly, the study revealed that 10% of the total contaminants appeared at the sites due to other anthropogenic or climatic factors. These include candy and sweet wrappers, residential litter as well as old vehicle spare parts. These and other plastic litter could be brought in due to environmental elements including wind and run-off which are critical in the movement and dispersal of macroplastics as reported by Kundu et al. (2021) and Liro et al. (2022). Also, the litter could come from neighboring roads and rural areas by wind or runoff; thus, non-agricultural contaminants are present in the parcels due to unlawful trash deposition.

However, the macroplastic litter in the area was characterized by different morphological structures, such as shape, size, color, and polymer composition. The findings of this research revealed that film was the dominant shape structure accounting for 74-95%. The transparent film was dominant in the areas because it is the same plastic shape and color that is used for greenhouse coverage. Likewise, previous studies reported macroplastic films as a major morpho-type found in greenhouse areas (Choi et al. 2020; Wang et al. 2022). Still, the polymer structure recovered in the areas was mainly PE which accounted for 79-93%. The reason for the high abundance of PE in the areas is that the chemical composition of covering materials (foil) is made up of PE materials. Also, some of the external macroplastic litter is made up of PE materials. Also, the findings reveal that contaminants in the litter are made up of different colors that originated from plastic materials used in the greenhouses such as foil, irrigation drip, threads, containers, creates, etc. Further, macroplastic litter from external sources contributed to the color sources in the greenhouse environment. The result of macroplastic litter revealed in this study can be valuable information to farmers and stakeholders for planning, cleaning, and dumping management. It could be relevant for plastic reused programs within the frame of the circular economy.

6.2 Microplastic pollution in the greenhouse soil surface and profiles

This study also aimed to assess microplastic pollution in the greenhouse soil, on the surface, and along soil profiles. The results indicated that microplastic contaminants pollute not just the surface, but also the sub-surface zones of greenhouse farmlands. An overall average of 440 pieces/kg of microplastic was found on the surface and sub-surface samples of the studied plots. The microplastic content on the soil surface (0-20 cm) was higher (mean: 475 pieces /kg) than that of the sub-soil (20-40 cm: mean: 338 pieces /kg). Thus, the microplastic contamination in the study area was similar to various studies that concluded that greenhouse farming is among the major contributor of microplastic in the environment (Choi et al. 2020; Isari et al. 2021: Wang et al. 2022). Yet, the result of this study also concluded that the differences in the microplastic contamination in the study areas could be reasoned by the still ongoing macroplastic weathering, the differences in agricultural practices, dissimilarities in abandoned duration, and probably the degree of cleaning the greenhouse films after the end of the agricultural activity.

Moreover, the findings indicate that the morphological structures found in the areas were mostly microplastic fiber (44%) and film (29%), as they probably originated from the fragmentation of macroplastic fiber materials and covering foil used in the greenhouse environment. In addition, the abundance of fiber could be traced to the deposition of fibrous materials by atmospheric deposition, water runoff, and wind (Rezaei et al. 2019; Allen et al. 2019; Haixin et al. 2022). The chemical composition of the contaminants found were PP (44%) and PE (29%). This can be traced to the fragmentation of macroplastic fiber and film used in the greenhouse environment. Similarly, the microplastic structures were mainly small in size, ranging between 0-1 mm (49%) and 1-2 mm (21%). The larger microplastic contaminants were recovered in the soil surface (0-20 cm), while the smaller microplastic contaminants were recovered in the soil sub-surface (20-40 cm). This occurred because the smaller microplastic contaminants such as fiber could easily go down to the soil depths through the soil pores and water infiltration, while the larger microplastic contaminants find it difficult to migrate to the soil depths. Transparent and blue colors were the dominant colors of the contaminants found in the areas. This result of morphological structure shape, size, and color was similar to what previous authors reported in the agricultural soils in different parts of the world (Piehl et al. 2018; Meng et al. 2020; Wang et al. 2022; Liu et al. 2022).

6.3 Vertical distribution of microplastic pollution in the soil profiles

This research aimed to evaluate the vertical distribution of microplastic pollution in the soil profiles. The overall mean abundance of microplastics in the soil profiles is 63.77± 14 pieces/kg, whereas in the surface layer, it was 440 pieces/kg. The analysis of variance shows that the abundance significantly differs in the different study areas, thus the means of the profiles were 85 pieces/kg at Szeged (on the surface: 225 ± 61pieces/kg), 4.5 pieces/kg at Szentes (on the surface: 125± 52 pieces/kg), and 96.3 pieces/kg at Szarvas (on the surface: 866± 102 pieces/kg). However, the contaminants in the soil profiles are mainly fibers (72%) and fragments (19%), whereas on the surface their proportions were 44 and 18% respectively. These structures were recovered in different colors, but transparent and red colors dominated the colors accounting for 37% and 23% respectively. So, we can conclude that less microplastic with special forms does migrate to the soil depths.

The results confirm that microplastic contaminated even the deeper layers of the soil.

However, the distribution of these contaminations is not uniform, but microplastics could be found in any layer, though, in the layer close to the surface higher contamination was recorded than in the deeper layers. The deepest layer where the microplastic contamination was recorded was 160 cm. Furthermore, the comparison between the microplastic pollution and the physical parameters of the soil samples confirms that soil texture and grain size distribution affect the vertical migration of microplastics in the soil profiles. Thus, microplastics were found to be more distributed in the silt loam than in the silty clay loam. Further, the results show that the migration of microplastics is not always influenced by particle distribution of the soil, but other physical parameters, such as soil porosity, bulk density, and hydraulic conductivity. These physical parameters influence the vertical migration of microplastics from one layer to another, thus, smaller microplastics could migrate downward, while the larger contaminants remained on the surface.

6.4 Processes of greenhouse film weathering and environmental contamination

Plastic greenhouse farming substantially relies on plastic materials for coverage, water piping, biding strings to support plants, crates for transportation, and other purposes. However, the plastic materials in the greenhouse environment have a short time. Decades ago, plastic films were manufactured to last 1–2 years. In recent years, with the advancement of technology, the durability and performance of these plastic films have increased by up to 3 years (Dehbi et al. 2015; Babagyayou et al. 2020). The findings of this study show that the weathering of greenhouse film and other plastic materials used in greenhouse farming is influenced by the action of several factors. The important factors include temperature, UV radiation, precipitation, agrochemicals, wind, atmospheric contamination, and greenhouse structure (Figure 27). For example, among the studied sites, the area of Szeged has the highest UV radiation, and temperature creating favorable climate conditions for the weathering of plastic materials. The combination of the parameters above adjusts the chemical composition of plastic materials, weakens the polymer structure, and causes depolymerization. The fragmented plastic particles end up in the soil as macroplastic litter. The macroplastic litter together with other non-agricultural contaminants that occurred in the greenhouse environment as a result of anthropogenic and climatic factors accumulate on the soil surface (Kundu et al. 2021; Liro et al 2022). The availability of macroplastic litter in the environment largely depends on the length of the greenhouse abandonment**,** the method and frequency of farm cleaning (Meng et al. 2020; Wang et al. 2022).

The macroplastic litter, however, experienced further weathering and fragmentation through additional factors such as farm management practices (eg. harrowing, plowing, and tilling). The fragmented plastic particles become plastic contaminants of different sizes. The smaller microplastic materials pollute the soil surface and migrate vertically into the soil and soil profiles influenced by different factors. This research found that the most important factors that influence this vertical migration include infiltration**,** clay, and sand content**,** similar to the results of Yu et al. (2022). Particle size distribution in the clay soils impedes the easy vertical migration of microplastic contaminants, contrarily, this study found that the particle size distribution in the sandy soils favors the easy movement of microplastic into deeper soil layers. Other factors that were found to influence the migration of microplastic contaminants include porosity, bulk density, and hydraulic conductivity of the soils (Oßmann, 2021; O'Connor et al., 2019; Schell et al., 2022). Soils with high porosity, low bulk density, and high hydraulic conductivity tend to have a high migration of microplastics. Furthermore, importantly, this study found that because of the nature of the soil texture and its hydraulic properties, the movement of these contaminants mostly occurred in the study areas through soil cracks**,** similar to the results of Cao et al. (2021). Both macro- and microplastic contaminants get into the cracks as a result of surface runoff, infiltration, wind action, living organisms, and tillage practices.

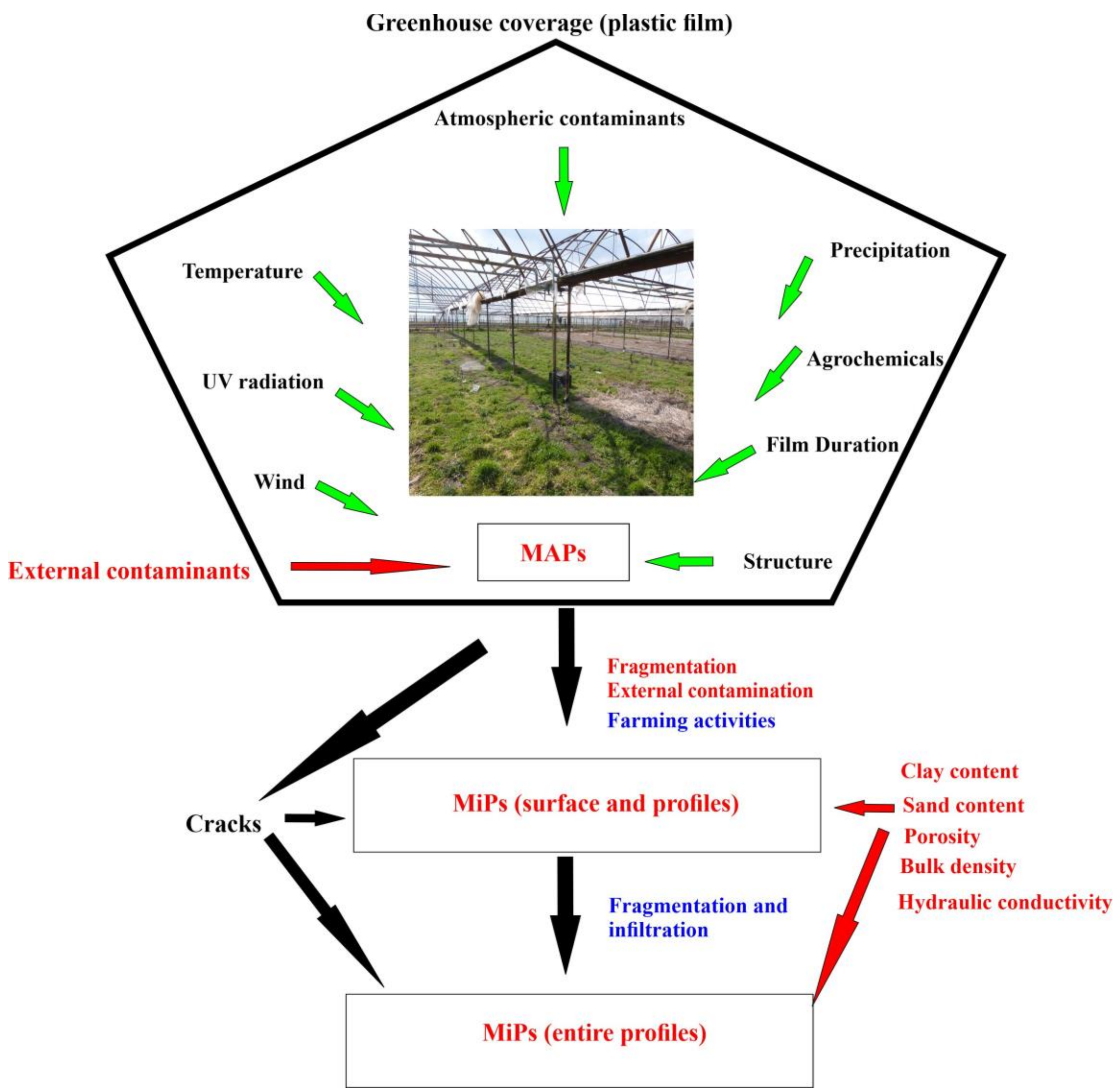

Figure 27: Conceptual model of greenhouse plastics fragmentation to microplastics and penetration to soil depths

6.5 Limitations of the Applied Methods

The limitation of this study lies in the difficulty in picking and purifying the macroplastic litter on the surface without causing damage or breakage. The research aimed to quantify the level of macroplastic litter on the surface. Breakage during the fieldwork and laboratory purification became one of the obstacles that affected this study. This might affect the number, shape, and size of macroplastic litter in the area. However, this problem was minimized by assigning careful observers during the fieldwork, and the macroplastic samples were gently handled during the laboratory analysis. Secondly, this research could not include the samples that were shallowly buried under the soil because the observers could only pick what was on the surface. This could affect the total amount of macroplastic litter in the area. Thirdly, the overstay of macroplastic litter on the soil together with the environmental conditions make some litters dirty and difficult to be identified as plastic contaminants. This may also affect the total number of macroplastic litter recovered in the areas.

Moreover, for microplastic analysis, this study could not analyze a large quantity of soil samples because of the difficulty in getting rid of organic matter in the soil as agricultural soils have a high content of organic matter. Thus, 10 g of soil samples were used in this study, and this could affect the generalizability of the findings. Also, environmental contamination during the laboratory was reported in this study (mostly fibers) despite the strict measures to prevent it. Even though this problem was detected by creating blank samples and subtracting the number of contaminants from the samples, this environmental contamination could affect the total number of contaminants in the soil samples.

6.6 Recommendations

Having mentioned the limitation of this study and given that macroplastics were found on the surface of abandoned greenhouse farmlands, and that these macroplastic litters fragment and form microplastics that contaminate the greenhouse surface and soil profiles, several recommendations could be formulated.

I recommend that (1) careful cleaning and disposal of plastics on greenhouse farmlands should be controlled by law, and the cleaning process should be observed. Furthermore, (2) prevention of greenhouse farmland contamination by external and nonagricultural litter due to anthropogenic and climatic factors is needed; thus, the farmers should be educated to eliminate or at least limit plastic pollution. Also (3) photographs of the sampling sites should be taken before the sample collections and later on the macroplastic litter recovered should be compared to determine the level of fragmentation due to the sample collection methods and laboratory analysis procedures. In addition (5) a $1m^3$ depth should be dug to collect the potential macroplastic litter that was buried possibly due to the effect of agricultural activities. The result obtained should be applied and determine the buried macroplastic litter in the entire farmland area. Further, (6) more samples should be used in future microplastic studies of Hungary together with standard microplastic extraction methods that can efficiently remove the organic matter from the high-quantity samples.

Comparative analysis of the two results (10g results and high-quantity samples results) should be made and the best result of the microplastic extraction method in Hungary should be chosen. On top of that (7), blank samples should be necessary to determine the level of environmental contamination as well as a method of correcting the level of environmental contamination from the samples. Also, (8) the preliminary information on the macroplastic litter is important as this could reveal the basic information of the morphological characteristics (such as colors, shapes, types, and polymer types) of the microplastic. This might ease the characterization of microplastic contaminants.

Further, this study recommends the following for future studies:

1. Obtaining enough samples that may yield more microplastic contaminants from the soils and be used for the generalizability of the findings.
2. Elucidate the duration of macroplastic fragmentation to microplastic contaminants in greenhouse environments.
3. Study of microplastics and nanoplastics contamination in the groundwaters. This is because the microplastics were found at depths very close to groundwater.
4. In-depth study of the contribution of soil cracks in the vertical migration of microplastic contaminants.
5. Perform further studies to shed light on the complex relationship between microplastic abundance and soil physical properties.

7. ACKNOWLEDGEMENTS

I would like to start my acknowledgment by thanking the almighty God who made it possible by sparing us life and good health to witness the successful end of this thesis write-up and completion of the Ph.D. program.

My sincere gratitude goes to my supervisors, Dr. Timea Kiss and the late Prof. Dr. Andrea Farsang, for their selfless guidance and training for this historical Ph.D. journey. For Dr. Timea Kiss, I owe you my deepest gratitude for coming to my supervision at a time of highest need. Your involvement in this work has reshaped it and added more value to this thesis. Also, your adequate supervision and training have molded me positively in time management, problem-solving, and perseverance in research activities. May your good wishes be granted and your selfless services to humanity give you everlasting happiness in your life. However, for the late Prof. Dr. Andrea Farsang of blessed memory, I thank you from the first day I contacted you until the day you left us in December 2022. Throughout our short stay, you not only acted as my supervisor and academic mentor but a mother who cared and was concerned about the progress of her son. The love you have shown to me and my family here in Szeged is memorable. Henceforth you will be regarded as part of my history.

Specifically, my deep appreciation goes to the members of my family, especially, my beloved mother, siblings, wife, and lovely daughter Aisha Ibrahim Saadu, for their constant support, prayers, and encouragement throughout my Ph.D. pursuit.

Special thanks to the Hungarian Government through the Tempus Public Foundation and the Federal Scholarship Board, Ministry of Education Nigeria for granting me the Stipendium Hungaricum scholarship and bilateral educational agreement scholarship which made my dream about studying abroad come true. I cherish your support, and I will always be a good example of the products of your great institutions.

I gratefully appreciate the entire academic and non-academic staff of the Department of Geoinformatics, Physical and Environmental Geography, for their constant guidance and positive contributions throughout the write-up of this thesis, academic pursuit, and my stay in the department. Indeed, I have learned a lot by being your student.

I want to especially recognize and thank Dr. Barta Károly, Mako Laszlo, and Gábor Gubucz, for helping me in carrying out the fieldwork of this study despite all the weather difficulties. Similarly, I would also thank Dr. Barta Károly, Dr. Fintor Krisztián, István

Fekete, and Dávid Filyó for helping me in carrying out the laboratory analysis. Thank you for being with me on this time-consuming exercise. Similar gratefulness goes to my Ph.D. colleagues who help me and always make me feel at home.

To the Nigerian community and my international friends, I am very much happy throughout our stay. I strongly believe that our departure from Szeged will only strengthen our relationship.

8. REFERENCES

Afrin, S., Uddin, M.K., Rahman, M.M. 2020. Microplastics contamination in the soil from urban landfill site, Dhaka, Bangladesh. Heliyon, 6/11, e05572.

Alhamdan, AM., Al-Helal, IM. 2009. Mechanical deterioration of polyethylene greenhouses covering under arid conditions. Journal of Materials Processing Technology, 206/1, 63–69.

Allen, S., Allen, D., Phoenix, VR., Roux, GL., Jiménez, PD., Simonneau, A., Binet, S., Galop, D. 2019. Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nat Geosci., 12, 339–344.

Andrady, AL. 2003. Plastics and the environment. John Wiley & Sons, Inc., New Jersey.

Astner, A.F., Hayes, DG., O'Neill, H., Evans, B.R., Pingali, S.V., Urban, V.S., Young, T.M. 2019. Mechanical formation of micro- and nano-plastic materials for environmental studies in agricultural ecosystems. Sci of the Total Environ., 685, 1097–1106.

Babagyayou, MI., Mourad, AI., Ochoa, A., Beltran, F., Cherupuraka, N. 2020. Study on the thermal stability of stabilized and unstabilized low-density polyethylene films. Polymer Bulletin. *78*/9, 5225-5241.

Barboza, LGA., Cózar, A., Gimenez, BCG., Barros, TL., Kershaw‖, PJ., Guilhermino, L. 2019. Macroplastics pollution in the marine environment. World Seas: an environmental evaluation (2nd Edition), 3, Ecological Issues and Environmental Impacts, 305-328.

Barnes, DKA., Galgani, F., Thompson, RC., Barlaz, M. 2009. Accumulation and fragmentation of plastic debris in global environments. Phil. Trans. R. Soc. B. 364, 1985–1998

Berenstein, G., Córdoba, P., Díaz, YB., González, N., Ponce, MB., Montserrat, JM. 2024. Macro meso, micro, and nanoplastics in horticultural soils in Argentina: Abundance, size distribution and fragmentation mechanism. Sci. of the Total Environ., 167672.

Berg, PV., Huerta-Lwanga, E., Corradini, F., Geissen, V. 2020. Sewage sludge application as a vehicle for microplastics in estern Spanish agricultural soils. Environmental Pollution, 261, 114198.

Beriot, N., Peek, J., Zornoza, R., Geissen, V., Huerta, LE. 2021. Low-density microplastics detected in sheep faeces and soil: a case study from the intensive vegetable farming in southeast Spain. Sci. of the Total Environ., 755,142653

Bhuyan, MS., 2022. Effects of Microplastics on Fish and in Human Health. Front. Environ. Sci., 10, 827289.

Bläsing, M., Amelung, W. 2018. Plastics in soil: analytical methods and possible sources. Sci Total Environ 612, 422–43.

Blocker, L., Watson, C., Wichern, F. 2020. Living in the plastic age - Different short-term microbial response to microplastics addition to arable soils with contrasting soil organic matter content and farm management legacy. Environmental Pollution 267, 115468.

Boots, B., Russell CW., Green DS. 2019. Effects of Microplastics in Soil Ecosystems: Above and Below Ground Environ. Sci. Technol., 53/19, 11496–11506.

Boughattas, I., Hattab, S., Zitouni, N., Mkhinini, M., Missawi, O., Bousserrhine, N., Banni, M., 2021. Assessing the presence of microplastic particles in Tunisian agriculture soils and their potential toxic changity effects using Eisenia andrei as bioindicator. Sci. Total Environ., 796, 148959.

Braun, M., Mail, M., Heyse, R., Amelung, W. 2020. Plastic in compost: prevalence and potential input into agricultural and horticultural soils. Sci Total Environ, 706, 143335.
Büks, F., Kaupenjohann, M. 2020. Global concentrations of microplastics in soils—a review. Soil, 6, 649–662.
Cao, L., Wu, D., Liu, P., Hu, W., Li, X., Sun, Y., Wu, Q., Tian, K., Huang, B., Yoon, SJ., Kwon, B., Khim, JS. 2021. Occurrence, distribution and afecting factors of microplastics in agricultural soils along the lower reaches of Yangtze River, China. Sci Total Environ., 148694.
Chen Y., Leng,Y., Liu, X., and Wang , J. 2020. Microplastic Pollution in Vegetable Farmlands of Suburb Wuhan, Central China. Environmental Pollution, 257, 113449.
Choi, YR., Kim, Y., Yoon, J., Dickson, N., Kim, K. 2020. Plastic contamination of forest, urban, and agricultural soils: a case study of Yeoju in the Republic of Korea. Journal of Soils and Sediments, 21/8, 1-12.
Contaminated Land Management Guidelines No. 5 Site Investigation and Analysis of Soils (Revised 2011). Published by the Ministry for the Environment Wellington, New Zealand
Corradini, F., Meza, P., Eguiluz, R., Casado, F., Huerta-Lwanga, E., Geissen, V. 2019. Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. Sci Total Environ., 671, 411–420
Crossman. J., Hurley, RR., Futter, M., Nizzetto, L. 2020. Transfer and transport of microplastics from biosolids to agricultural soils and the wider environment. Sci Total Environ., 724, 138334.
Cyvin, JB., Ervik, H., Kveberg, AA., Hellevik, S. 2021. Macroplastic in soil and peat. A case study from the remote islands of Mausund and Froan landscape conservation area, Norway; implications for coastal cleanups and biodiversity. Science of the Total Environ., 787. 147547
Das, A., and Guo, H. 2023. Raman Spectroscopy, Encyclopedia of Soils in the Environment (Second Edition), 2, 497-509.
Dehbi, A., Mourad, AI., Djakhdane, K., Hilal-Alnaqbi, A. 2015. Degradation of thermomechanical performance and lifetime estimation of multilayer greenhouse polyethylene films under simulated climatic conditions. Polymer Engineering and Science 55/2, 287-298.
Dilara, PA., Briassoulis, D. 2000. Degradation and stabilization of low-density polyethylene film used as greenhouse covering materials. J. Agric. Engng. Res., 76, 309-321.
Ding, L., Wang, X., Yang, X., Zhang, C., Qi, Y., Guo, X. 2020. The occurrence and distribution characteristics of microplastics in the agricultural soils of Shaanxi Province, in north-western China. Sci. of the Total Environ. 720/10,137525.
Dövényi. Z. 2010. Microregions of Hungary (in Hungarian) MTA FKI. Budapest, Hungary, 281–287
Du, C., Liang H., Li Z., Gong J. 2020. Pollution characteristics of microplastics in soils in southern suburbs of Baoding City China. International Journal of Environmental Research and Public Health. 17, 845.
Enders, K., Lenz, R., Ivar, J.A., Tagg, A.S., Labrenz, M. 2020. When every particle matters: A QuEChERS approach to extract microplastics from environmental samples. MethodsX, 7, 100784.

Enyoh, CE., Verla, AW., Verla, EV., Enyoh, EC. 2020. Effect of macro- and microplastics in soil on quantitative phytochemicals in a different part of juvenile lime tree (Citrus aurantium). AIMS Environmental Science, 7/6, 526–541.

Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S. 2013. Microplastic pollution in the surface waters of the Laurentian Great Lakes. Mar. Pollut. Bull., 77, 177e182.

Faix, O. 1992. Fourier transform infrared spectroscopy. Springer Verlag Berlin Heidelberg, Berlin

Fakour, H., Lo, SL, Yoashi, NT., Massao, AM., Lema, NN., Mkhontfo, FB., Jomalema, PC., Jumanne, NS., Mbuya, BH., Mtweve, JT., Imani, M. 2021. Quantification and analysis of microplastics in farmland soils: characterization, sources, and pathways. Agriculture, 11/4, 11040330

Filep, G. 1998. Soil Pollution. Rexpo Ltd Debrecen, Hungary.

Giacomelli, GA. and Roberts, WJ. 1993. Greenhouse covering system. Horttechnology 3/1, 50–58.

Gies, EA., LeNoblea, JL., Noëla, M., Etemadifara, A., Bishayc, F., Hallb, ER., Ross, PS. 2018. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. Mar. Pollut Bull., 133, 553–55.

Grause, G., Kuniyasu, Y., Chien, M., Inoue, C. 2022. Separation of microplastic from soil by centrifugation and its application to agricultural soil. Chemosphere. 288/3, 132654.

Haixin, Z., Yimei, H., Shaoshan, A., Haohao, L., Xiaoqian, D., Pan, W., Mengyuan, F., 2022. Land-use patterns determine the distribution of soil microplastics in typical agricultural areas on the eastern Qinghai-Tibetan Plateau. Journal of Hazardous Materials. 426, 127806.

Hamid, FS., Bhatti, MS., Anuar, N., Anuar, N., Mohan, P., Periathamby, A. 2018. Worldwide distribution and abundance of microplastic: How dire is the situation? Waste Management & Research, 36/10, 873–897.

Harms, IK., Diekötter, T., Troegel, S., Lenz, M. 2021. Amount, distribution, and composition of large microplastics in typical agricultural soils in northern Germany. Sci. Total Environ., 758. 143615.

He, D., Luo, Y., Lu, S., Liu, M., Song, Y., Lei, L. 2018. Microplastics in soils: Analytical methods, pollution characteristics, and ecological risks. Trends in Analytical Chemistry 109, 163-172.

Hernández-Gutiérrez, E., Osten, JR., Escalona-Segura, G., Mendoza-Vega, J., Dzul-Caamal, R., Posthumus, S., Vastenhouw, R., Yang, X., Geissen, V., Huerta-Lwanga, E. 2022. Morphospecies abundance of above-ground invertebrates in agricultural systems under glyphosate and microplastics in south-eastern Mexico. *Environments, 8*/11, 130.

Hidalgo-Ruz, V., Gutow, L., Thompson, RC., Thiel, M. 2012. Microplastics in the marine environment: A review of the methods used for identification and quantification. Environmental Sciences & Technology. 46, 3060−3075.

Hidayaturrahman, H., Lee, T. 2019. A study on characteristics of microplastic in wastewater of South Korea: Identification, quantification, and fate of microplastics during treatment process. Marine Pollution Bulletin, 146, 696-702.

Hu, Q., Xianyue, Li, X., Gonçalves, JM., Shi, H., Tian, T., Chen, N. 2020. Effects of residual plastic-film mulch on field corn growth and productivity. Science of the Total Environ, 729. 138901.

Huang, C.Y., Xu, J.M., 2010. Soil Science. 3rd edition. China Agriculture Press, Beijing, China (In Chinese).
Huang, Y., Liu, Q., Jia, W., Yan, W., Wang, J. 2020. Agricultural plastic mulching as a source of microplastics in the terrestrial environment. Environmental Pollution. 260.
Hurley, RR., Lusher, AL., Olsen, M., Nizzetto, L. 2018. Validation of a method for extracting microplastics from complex, organicerich, environmental matrices, Environ. Sci. Technol., 52, 7409e7417
Isari, EA., Papaioannou, D., Ioannis, K., Kalavrouziotis, IK., Karapanagioti, HK. 2021. Microplastics in agricultural soils: a case study in cultivation of watermelons and canning tomatoes. *Water, 13/16,* 2168.
Ivar do Sul, J.A. and Costa, M F. 2014. The present and future of microplastic pollution in the marine environment. Environmental Pollution. 185. 352-364.
Jiang, S., Wang , F., Li, Q., Sun, H., Wang, H., Yao, Z. 2021. Environmental and food safety: a novel integrative review. Environ Sci. Pollut. Res. Int., 28/39, 54511-54530.
Julienne, F., Delorme, N., Lagarde, F. 2019. From macroplastics to microplastics: role of water in the fragmentation of polyethylene. Chemosphere, 236, 124409.
Kalogerakis, N., Karkanorachaki, K., Kalogerakis, GC., Triantafyllidi, EI., Gotsis. AD., Partsinevelos, P., Fava, F. 2017. Microplastics generation: onset of fragmentation of polyethylene films in marine environment mesocosms. Front. Mar. Sci., 84/4, 00084.
Katsumi, N., Kusube, T., Nagao, S., Okochi, H. 2020. Accumulation of microcapsules derived from coated fertilizer in paddy fields. Chemosphere, 267,129185.
Kawecki, D., Nowack, B. 2021. A proxy-based approach to predict spatially resolved emissions of macro and microplastic to the environment. Sci Total Environ, 15/748. 141137.
Kim, S., Kim, J., Lee, H., Lee, H. 2021. Abundance and characteristics of microplastics in soils with different agricultural practices: Importance of sources with internal origin and environmental fate. Journal of Hazardous Materials, 403, 123997.
Kormany.hu: New period in the recycling of plastic films with agricultural origin (In Hungarian) https://kormany.hu/hirek/uj-korszak-a-mezogazdasagi-foliahulladekok-ujrahasznositasaban. (Assessed on 25/03/2023)
KSH (National Regional Development and Spatial Planning Information System), County-scale data on plastic and glass greenhouse areas between 2011 and 2022. (in Hungarian) (Assessed on 28. April 2023)
Kumar, VM., Sheela, MA. 2021. Effect of plastic film mulching on the distribution of plastic residues in agricultural fields. Chemosphere, 273, 128590.
Kun, A., Katona, O., Sipos, G., Barta, K. 2013. Comparison of pipette and laser diffraction methods in determining the granulometric content of fluvial sediment samples. Journal of Environmental Geography 6/4, 49–54.
Kundu, MN., Komakech, HC., Lugomela, G. 2021. Analysis of macro- and microplastics in riverine, riverbanks, and irrigated farms in Arusha, Tanzania. Archives of Environmental Contamination and Toxicology. Archives of Environmental Contamination and Toxicology 82/3, 142-157.
Lechthaler, S., Waldschläger, K., Stauch, G., Schüttrumpf, H. 2020. The Way of Macroplastic through the Environment. Environments, 7/10, 73.
Lenz, R., Enders, K., Stedmon, CA., Mackenzie, DMA., Nielsen, TG. 2015. A critical assessment of visual identification of marine microplastic using Raman spectroscopy

for analysis improvement, Mar. Pollut. Bull., 100, 82-91.
Li, L., Luo, Y., Li, R., Zhou, Q., Peijnenburg, WJ. Yin, N., Yang, J., Zhang, YC. 2020. Effective uptake of submicrometre plastics by crop plants via a crack-entry mode. Nature Sustainability, 3, 929 – 937.
Li, Q., Wu, J., Zhao, X., Gu, X., Ji, R. 2019. Separation and identification of microplastics from soil and sewage sludge. Environ Pollut., 254(Part B), 113076.
Li, S., Ding, F., Flury, M., Wang, Z., Xu, L., Li, S., Jones, DL., Wang, J. 2022. Macro- and microplastic accumulation in the soil after 32 years of plastic film mulching. Environmental Pollution, 300, 118945.
Liro, M., Emmerik, THM., Zielonka, VA., Gallitelli, L., Mihai, F. 2023. The unknown fate of macroplastic in mountain rivers. Sci. of the Total Environ., 865, 161224
Liro, M., Mikuś, P., Wyżga, B. 2022. First insight into the macroplastic storage in a mountain river: the role of in-river vegetation cover, wood jams, and channel morphology. Sci. Total Environ., 838, 156354.
Liu, M., Lu, S., Song, Y., Lei, L., Hu, J., Lv, W., Zhou, W., Cao, C., Shi, H., Yang, X., He, D. 2018. Microplastic and mesoplastic pollution in farmland soils in suburbs of Shanghai, China. Environ Pollution 242/B, 855–862.
Liu, X., Tang, N, Yang, W., Chang, J. 2022. Microplastics pollution in the soils of various land-use types along Sheshui River basin of Central China. Sci. of the Total Environ., 806/3, 150620
Lu, Y., Zhang, JD., Liu, Y., Chen, LY., Tao, S., Liu, WX. 2021. Distribution characteristics of microplastics in agricultural soils from the largest vegetable production base in China. Sci. of the Total Environ., 756, 143860.
Luqman, A., Nugrahapraja, H., Wahyuono, R.A., Islami, I., Haekal, M.H., Fardiansyah, Y., Putri, BQ., Amalludin, F.I., Rofiqa, E.A., Götz, F., Wibowo AT. 2021. Microplastic contamination in human stools, foods, and drinking water associated with Indonesian coastal population. *Environments, 8*, 138.
Lusher, A., Hurley, R., Vogelsang, C., Nizetto, L. 2018. Mapping microplastics in sludge. Norwegian Institute for Water Research Technical Report. 7215, 1–55.
Maaß, S., Daphi, D., Lehmann, A., Rillig, MC. 2017. Transport of microplastics by two 556 collembolan species. Environmental Pollution, 225, 557, 456-459.
Machado, AA., Lau, CW., Till, J., Kloas, W., Lehmann, A., Becker, R., Rillig, MC. 2018. Impacts of Microplastics on the Soil Biophysical Environment. Environ. Sci. Technol., 52/17, 9656–9665.
Markovi,´ S.B., Vandenberghe, J., Peri´c, Z.M., Filyó, D., Bartyik, T., Radakovi´c, M.G., Hao, Q,, Markovi´c, R.S., Luki´c, T., Tomi´c, N., Gavrilov, M.B., Anti´c, A., Cvijanovi´c, I., Sipos, G., 2023. Local differentiation in the loess deposition as a function of dust source: key study novo orahovo loess paleosol sequence (Vojvodina, Serbia). Quaternary, 6/1, 23.
Meides, N., Mauel, A., Menzel, T., Altstädt, V., Ruckdäschel, H., Senker, J., Strohriegl, P., 2022. Quantifying the fragmentation of polypropylene upon exposure to accelerated weathering. Environ. Sci. Europe, 2: 23.
Meixner, K., Kubiczek, M., Fritz, I., 2020. Microplastic in soil–current status in Europe with special focus on method tests with Austrian samples. AIMS Environ. Science, 7/2, 174–191.

Meng, F., Fan, T., Yang, X., Riksen, M., Xu, M., Geissen, V. 2020. Effects of plastic mulching on the accumulation and distribution of macro and micro plastics in soils of two farming systems in Northwest China. PeerJ 8. e10375.

MERI. Guide to Microplastic Identification; Marine and Environmental Research Institute: Blue Hill, ME, USA, 2017; p. 15.

Mo, X., Li, H., Lian, Y., Zheng, B., Dong, J., Lu, X. 2021. Estimation of soil microplastic input derived from plastic gauze using a simplified model. Sci Total Environ, 798, 148577.

Mora, A., García-Gamboa, M., Sánchez-Luna, MS., Gloria-García, L., Cervantes-Avilés, P., Mahlknecht, J. 2021. A review of the current environmental status and human health implications of one of the most polluted rivers of Mexico: The Atoyac River, Puebla. Sci of the Total Environ., 782, 146788.

Németh, Sz., and Ehret-Berczi, I., 2014. The Hungarian horticultural sector: Economic analysis of tomato greenhouse farm. Acta Horticulturae, 1041, 307– 310.

Nuelle, M.T., Dekiff, J.H., Remy, D., Fries, E., 2014. A new analytical approach for monitoring microplastics in marine sediments. Environ. Pollut., 184, 161–169.

O'connor, D., Pan. S., Shen. Z., Song. Y., Jin. Y., Wu. W., Hou. D. 2019. Microplastics undergo accelerated vertical migration in sand soil due to small size and wet-dry cycles. Environ. Pollut. 249,527–534.

Oßmann, BE., 2021. Microplastics in drinking water? Present state of knowledge and open questions. Current Opinion in Food Science. 41, 44-51.

Patel, A., Tandel, Y. 2017. Use of plastics in horticulture production. Indian Farmer 4/3,108-112.

Pazienza, P., Lucia, CD. 2020. For a new plastics economy in agriculture: policy reflections on the EU strategy from a local perspective. J Clean Prod.

Peng, G., Xu, P., Zhu, B., Bai, M., Li, D. 2018. Microplastics in freshwater river sediments in Shanghai, China: A case study of risk assessment in mega-cities. Environ. Pollut., 234,448-456.

Piehl, S., Leibner, A., Löder, MGJ., Dris, R., Bogner, C., Laforsch, C. 2018. Identification and quantification of macro- and microplastics on agricultural farmland. Scientific Reports 8/1, 17950.

Pietz, O., Augenstein, M., Georgakakos, CB., Singh, K., McDonald, M., Walter, MT. 2021. Macroplastic accumulation in roadside ditches of New York State's Finger Lakes region (USA) across land uses and the COVID-19 pandemic. Journal of Environ. Mgt., 298. 113524.

Plastic Europe. 2019. Plastic the facts. An analysis of European plastic production, demand and waste data. http://www.plasticeurope.org/application/file/9715/71299584/FINAL_web_version_plastics_the facts2019_14102019.pdf

Plastic Europe. 2020. Plastic the facts. An analysis of European plastic production, demand, and waste data. https://plasticseurope.org/knowledge-hub/plastics-the-facts-2020/

Plastic Europe. 2022. Plastic the facts. An analysis of European plastic production, demand, and waste data. https://plasticseurope.org/knowledge-hub/plastics-the-facts-2022/

Plastic Europe. 2023. Plastic the facts. An analysis of European plastic production, demand, and waste data. https://plasticseurope.org/knowledge-hub/plastics-the-fast-facts-2023/

Qi, Y., Yang, X., Pelaez, AM., Lwanga, HE., Beriot, N., Gertsen, H., Garbeva, P., Geissen,

V. 2018. Macro- and micro-plastics in soil-plant system: Effects of plastic mulch film residues on wheat (Triticum aestivum) growth. Sci. of the Total Environ., 645, 1048–1056.
Ragoobur, D., Huerta-Lwanga, E., Somaroo, G.D. 2021. Microplastics in agricultural soils, wastewater effluents and sewage sludge in Mauritius. Sci Total Environ., 798, 149326.
Ramos, L., Berenstein, G., Hughes, EA., Zalts, A., Montserrat, JM. 2015. Polyethylene film incorporation into the horticultural soil of small periurban production units in Argentina. Sci. of the Total Environ, 523, 74-81.
Rezaei, M., Riksen, MJPM., Sirjani, E., Sameni, A., Geissen, V. 2019. Wind erosion as a driver for transport of light density microplastics. Sci. of the Total Environ., 669, 273–281.
Saadu, I. and Farsang, A. 2022. Greenhouse farming as a source of macroplastic and microplastics contamination in agricultural soils: A case study from Southeast Hungary. Agrochemistry and Soil Science, 71/1, 43-57.
Saadu, I., Farsang, A. 2023. Plastic contamination in agricultural soils: a review. Environ. Sci. Europe, 35, 63.
Sayadi-Gmada, S., Rodriguez-Pleguezuelo, CR., Rojas-Serrano, F., Parra-Lopez, C., Parra-Gomez, S., Garcia-Garcia, MC., Lorbach-Kelle, MB., Manrique-Gordillo, T. 2019. Inorganic waste management in greenhouse agriculture in Almeria (SE Spain): towards a circular system in intensive horticultural production. Sustainability, 11/14, 3782.
Schell, T., Hurley, R., Buenaventura, NT., Mauri, PV., Nizzetto, L., Rico, A., Vighi, M., 2022. Fate of microplastics in agricultural soils amended with sewage sludge: Is surface water runoff a relevant environmental pathway? Environmental Pollution, 293, 118520.
Scheurer, M., Bigalke, M., 2018. Microplastics in Swiss floodplain soils. Environ. Sci. Technol. 52/6, 3591–3598.
Schlotter, NE., 1989. Raman Spectroscopy. Comprehensive Polymer Science and Supplements, 1, 469-497.
Schothorst, B., Beriot, N., Lwanga, EH., Geissen, V. 2021. Sources of light-density microplastic related to two agricultural practices: the use of compost and plastic mulch. Environments 8/4, 36.
Stefano, N., Pleissner, D., 2022. Quantification and analysis of surface macroplastic contamination on arable areas. Journal of Soils and Sediments, 22, 757–768.
Steinmetz, Z., Löffler, P., Eichhöfer, S., David, J., Muñoz, K., Schaumann, G.E. 2022. Are agricultural plastic covers a source of plastic debris in soil? A first screening study. SOIL, 8, 31–47.
Su, S., Zhou, S., Lin, G., 2021. Existence of microplastics in soil and groundwater in Jiaodong peninsula. E3S Web of Conferences, 251, 02045.
Susanna, G. 2018. Plastic pollution in soil. Interactive soil quality assessment. Institute for european environmental policy waste in Switzerland. Soil Use Manage. 1–12. https://ieep.eu/uploads/articles/attachments/3a12ecc3-7d09-4e41-b67cb8350b5ae619/Plastic%20pollution%20in%20soil.pdf?v=63695425214
Svedin, J. 2020. Photodegradation of macroplastics to microplastics: a laboratory study on common litter found in urban areas. Unpublished master thesis at the department of natural resource engineering, Luleå University of Technology.
Szolnoki, Z., Farsang, A.,Pus kás I., 2013. Cumulative impacts of human activities on urban garden soils: origin and accumulation of metals. Environmental Pollution, 177, 106–

115.

Tagg, AS., Brandes, E., Fischer, F., Fischer D., Brandt J., Labrenz M. 2022. Agricultural application of microplastic-rich sewage sludge leads to further uncontrolled contamination. Sci Total Environ, 806(Pt 4), 150611.
Thomas, D., Schütze, B., Heinze, WM., Steinmetz Z. 2020. Sample preparation techniques for the analysis of microplastics in soil—a review. Sustainability, 12, 9074.
Vaughan, R., Turner, SD., Rose, NL. 2017. Microplastics in the sediments of a UK urban lake. Environ Pollut., 229, 10-18.
Veerasingam, S., Mugilarasam, M., Venkatachalapathy, R. 2020. Infuence of 2015 food on the distribution and occurrence of microplastic pellets along the Chennai coast, India. Mar. Pollution Bull, 109, 196–204.
Vermeiren, P., Muñoz, C., Ikejima, K. 2020. Microplastic identification and quantification from organic-rich sediments: A validated laboratory protocol. Environ. Pollution, 262, 114298.
Vox. G., Schettini, E., Stefan, L., Modesti, M. 2008. Effects of agrochemicals on the mechanical properties of plastic films for greenhouse covering. Acta Horticulturae 801/801, 155-162.
Wang, F., Zhang, X., Zhang, S., Zhang, S., Sun, Y. 2020. Interactions of microplastics and cadmium on plant growth and arbuscular mycorrhizal fungal communities in agricultural soil. Chemosphere, 254, 126791.
Wang, J., Coffin, S., Sun, C., Schlenk, D., Gan, J. 2019. Negligible effects of microplastics on animal fitness and HOC bioaccumulation in earthworm Eisenia fetida in soil. Environ Pollution. 249, 776-784.
Wang, K., Chen, W., Tian, J., Niu, F., Xing, Y., Wu, Y., Zhang, R., Zheng, J., Xu, L. 2022. Accumulation of microplastics in greenhouse soil after long-term plastic film mulching in Beijing, China. Sci of the Total Environ., 1, 828, 154544.
Watteau, F., Dignac, M., Bouchard, A., Revallier, A., Houot, S. 2018. Microplastic detection in soil amended with municipal solid waste composts as revealed by transmission electronic microscopy and pyrolysis/gc/ms. Front. Sustain. Food Syst., 2, 81.
Weinstein, JE., Crocker, BK., Gray, AD. 2016. Plastic debris in the aquatic environment from macroplastic to microplastic: degradation of high-density polyethylene, polypropylene, and polystyrene in a salt marsh habitat. Environmental Toxicology and Chemistry, 35, 7, 1632–1640.
Wittwer, SH. 1993. World-wide use of plastics in horticultural production. Hort. Tech 3/1, 6-19.
Wu, R., Cai, Y., Xing, S., Yang, Y., Mi, J., Liao, X. 2021. A Novel Method for Extraction of Polypropylene Microplastics In Swine Manure. Environ Scie and Pollution Research, 28, 13021–13030.
Wu, X., Liu, Y., Yin, S., Xiao, K., Xiong, Q., Bian, S., Liang, S., Hou, H., Hu, J., Yang, J. 2020. Metabolomics revealing the response of rice (Oryza sativa L.) exposed to polystyrene microplastics. Environ Pollution, 266, 115159.
Yang, M., Huang, D., Tian, Y., Zhu, Q., Zhang, Q., Zhu, H., Xu, C., 2021. Influences of different source microplastics with different particle sizes and application rates on soil properties and growth of Chinese cabbage (Brassica Chinensis L.). Ecotoxicology and Environmental Safety, 222, 112480.
Yu, L., Zhang, JD., Liu, Y., Chen, LY., Tao, S., Liu, LW. 2021. Distribution characteristics

of microplastics in agricultural soils from the largest vegetable production base in China. Science of the Total Environment 756. 143860.
Yu, Z., Song, S., Xu, X, Ma, Q., Lu, Y. 2021. Sources, migration, accumulation and influence of microplastics in terrestrial plant communities. Environ Exp. Bot., 192, 104635.
Zhang, GS., Liu, YF. 2018. The distribution of microplastics in soil aggregate fractions in southwestern China**.** Sci Total Environ. 15/642,12-20.
Zhang, M., Zhao, Y., Qin, X., Jia, W., Chai, L., Huang, M., Huang, Y. 2019. Macroplastics from mulching film is a distinct habitat for bacteria in farmland soil. Sci Total Environ., 20,688, 470-478.
Zhang, X., Chen, Y., Li, X., Zhang, Y. Gao, W., Jiang, J., Mo, A., He, D. 2022. Size/shape-dependent migration of microplastics in agricultural soil under simulative and natural rainfall. Sci of the Total Environ., 815, 152507.
Zhao, Z., Zhao, K., Zhang, T., Xu, Y., Chen, R., Xue, S., Liu, M., Tang, D., Yang, X., Giessen, V. 2022. Irrigation-facilitated low-density polyethylene microplastic vertical transport along soil profile: An empirical model developed by column experiment. Ecotoxicology and Environmental Safety, 247/12, 114232.

9. ABSTRACT

Plastic materials play a vital role in the greenhouse environment, as they are used not only in the structure of the greenhouses but also during agricultural production until post-harvest. Extensive greenhouse farming generates macro and microplastic waste in large quantities and pollutes farmlands in various environments. Especially greenhouse plastic cover is problematic, as it is weathered into larger macroplastic litter, and further fragmented into microplastic particles which contaminate the soil surface and migrate vertically into the soil layers.

Despite the contribution of greenhouse farming in the generation of plastic contaminants in the environment, there is a paucity of knowledge on the level of plastic contamination as well as the nature of macroplastic fragmentation to microplastics in the greenhouse environment. Recent studies tried to quantify the level of plastic pollution in the greenhouse environment, but their effort was mainly concentrated on the microplastic in the soil surface, so few studies that were conducted were concentrated on the soil surface layers. Thus, there is a paucity of information about the macroplastic contamination in the greenhouse farmlands. Also, there is still unexplored information about the level of microplastic pollution in the soil profile beyond 100 cm depth as well as the influence of soil physical properties on the soil migration.

Therefore, the 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) describing the

morphological structures of microplastic pollutants in the profiles, and (e) examining the influence of soil physical parameters on the distribution of microplastics.
Within the frame of this study, three abandoned greenhouse farmlands were selected, where I quantified the level of macro- and microplastic contamination.

During the fieldwork, visible macroplastic materials were picked by careful observers from the surface of the sample parcels of the greenhouse farmlands. For microplastic samples, 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) and the composite of four samples was made. Drills were made in the middle of parcels and the soil profile samples were taken at 20, 40, and 50 cm intervals.

In the laboratory, to purify the macroplastic, the plastic items were placed in buckets filled with tap water for 48 hours. Later, the macroplastic litter was air-dried and categorized into sizes, shapes, colors, polymer composition, and types. The microplastic was extracted from the soil by oven-drying (40°C), gently grinding into smaller pieces, and sieving with a 5 mm sieve. Digestion of soil organic matter was performed using 30% H_2O_2, and Fenton reagent 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 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, and later to Raman spectroscopy for polymer identification.

The mean macroplastic abundance was 2655 pieces/ha, which is equivalent to 5 kg/ha of macroplastic scattered on the surface of the studied greenhouse farmlands. In terms of number, the maximum mean abundance (4328 pieces/ha) was found in Szeged, and the second-highest abundance (3513 pieces/ha) was found in Szarvas. The site at Szentes was the least polluted (125 pieces/ha). Likewise in terms of weight, the highest contamination was found in Szarvas (10.68 kg/ha), followed by Szeged (6.4 kg/ha) and Szentes site (1.17 kg/ha); thus, the field at Szarvas and Szeged were 9 and 5 times more contaminated than the abandoned greenhouse at Szentes. The reasons for the variation of the macroplastic litter abundance in the areas could be the differences in greenhouse farming and farmland abandonment duration, clearing activities, and climate. The extended duration (27 years) of greenhouse farming in Szarvas may be the reason for the highest litter weight contamination.

The most prevalent size of the macro contaminants was 1-5 cm, and the fragmentation of microplastic contaminants was found to be an ongoing process in most analyzed areas. Different macroplastic structures were found, but film structure was the dominant accounting for 74-95% of the total contaminants. A transparent color, which is mostly the color of the film cover of the greenhouse, was found to be the most abundant accounting for 68%. Similarly, the polymer composition results show that 79-93 % of the macroplastic material recovered in the areas was made up of PE. These results confirmed that the macroplastic recovered in the study sites were made up of plastic materials used for mainly greenhouse coverage. The morphological results of the contaminants also 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. However, the results importantly show that not all the macroplastic materials found on the surface of greenhouse farmlands have an agricultural origin. Only 90% of litter was confirmed to be agricultural-related, while 10% was confirmed to originate from external sources due to the influence of wind, water runoff, and anthropogenic factors.

The results reflect that the microplastic contaminated the soil surface and sub-surface. The average microplastic abundance in the entire study area was 440 pieces/kg. The highest contamination was recorded in Szarvas fallow farmlands with an average abundance of 866±102 pieces/kg, while the least abundance was recorded in Szentes with 125±52 pieces/kg. The results reveal that the distribution of microplastic contaminants is not uniform in depth: as the soil surface (0–20 cm) is more polluted by microplastics (300±93 pieces/kg) than the sub-surface (20–40 cm: 150 ± 76 pieces/kg).

The morphological characteristics of the contaminants show that the microplastic recovered in the soil surface and sub-surface had different sizes of which 0-1 mm was the predominant size accounting for 49% of the particles. Larger microplastic contaminants were recovered in the soil surface (0-20 cm), while the smaller microplastic contaminants were recovered in the soil sub-surface (20-40 cm). Furthermore, of the different shapes recorded in the study areas fibers were the most predominant structure (44%) found in the entire area. 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. Considering the color, transparent color was found to be more abundant accounting for 35%. The results of the overall frequency of the polymer types reflect that PP was the dominant polymer (44%). The result polymer composition and color 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.

The result of microplastic distribution in the soil profiles shows that microplastic pollutes different layers of soil profiles. The deepest contamination was found in 160 cm. The average microplastic abundance varied among the analyzed profiles: 80 pieces/kg (Szeged), 4 pieces/kg (Szentes), and 96 pieces/kg (Szarvas). However, microplastic appeared at the greatest depths (160 cm) in Szeged, whereas their deepest appearance was at 50 cm in Szentes, and 140 cm in Szarvas. The results also show that the particle size distribution of the soil is a very important parameter in determining the distribution and migration of plastic pollutants in the soil and soil profiles. 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 fine silt (3.9-7.8 µm) soil samples. A negative correlation existed between microplastic abundance and clay contents. The morphological type of contaminants found in the soil depths was mostly fiber of smaller sizes with distinctive transparent colors.

Similarly, the results show that soil physical parameters may affect the distribution of microplastics at various depths in the soil profiles. The result confirmed that high soil porosity and bulk density affected the rate of microplastic migration in the soil profiles, while the hydraulic conductivity impeded the rate of microplastic migration in the soil profiles 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). Generally, the contribution of the aforementioned soil physical parameters was found to be minimal on the level of microplastic migration, especially in the soil depths and layers with high clay content. Thus, the result confirmed that the migration of microplastic contaminants may be probably due to the influence of soil cracks.

Having macroplastic and microplastic contaminants in the greenhouse agricultural environment, this study recommended the formulation of laws that will compel the careful cleaning and disposal of plastic remnants in the greenhouse environment as well as the prevention of greenhouse farmland contamination by external and nonagricultural litter due to anthropogenic and climatic factors. Lastly, the study highlighted and shed light on some areas that need future studies. These include (1) the duration of macroplastic fragmentation to microplastic contaminants in greenhouse environments, (2) study of microplastics and nanoplastic contamination in the groundwaters, (3) in-depth study of the contribution of soil cracks in the vertical migration of microplastic contaminants, and (4) study of the complex relationship between microplastic abundance and soil physical properties.

Statement of the supervisor

I, Dr. Tímea Kiss, as supervisor, declare that the thesis written by Saadu Ibrahim (Neptun code: OZ0OK5) titled Quantification of macro-and microplastic pollution of fallow greenhouse farmlands: case study in southeastern Hungary is his own writing prepared under my supervision; the candidate's contribution to the results used in the discussion of the thesis is approved. I also declare that the thesis meets the formal and professional requirements of the Doctoral School of Geosciences of the University of Szeged and the Faculty of Science and Informatics/ Department of Geoinformatics, Physical, and Environmental Geography; thus, I support its submission.

Date: 11/01/2024
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UNIVERSITY OF SZEGED
Faculty of Sciense and Informatics
Department of Geoinformatics, Physical and Environmental Geography
SZEGED
2 Egyetem str., Szeged H-6722

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Supervisor