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An In-Depth Analysis of Physical, Chemical, and Microplastic Parameters of Landfill Fine Fraction for Biocover Construction

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Abstract: Landfills pose global challenges, notably in terms of greenhouse gas (GHG) emissions, pollution release, and extensive land occupation. The transformative practice of landfill mining has redefined these sites as valuable resource reservoirs. The fine fraction (FF), often constituting the majority of excavated waste, is currently underutilized but holds the potential for biocover construction to mitigate methane emissions. This study comprehensively analyzes the FF from the Kuršenai landfill, collecting samples from various depths, reaching up to 10.5 m. The most suitable layers for biocover construction were determined based on basic physical and chemical parameters, along with the concentration of heavy metals and microplastics. The findings unveil significant parameter variations across different depths. Moderate–high correlations (ranging from 0.5 to 0.84) between several parameters were observed. The layer at a depth of 4.5–6 m emerged as the most suitable for biocover construction. However, this layer is characterized by elevated microplastic concentrations (30,208 ± 273 particles/kg), posing a challenge for its use in biocovers as microplastics can be released into the environment during FF extraction and biocover construction. Additionally, microplastics become finer with depth, increasing the associated risks. Therefore, a balanced approach considering material properties and pollution concentrations is vital for sustainable waste management practices.

Keywords: landfills; biocover; fine fraction; microplastics; methane; landfill mining

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1. Introduction

Landfills have long occupied significant areas in Europe, serving as repositories of waste materials that represent both a challenge and an opportunity. Europe hosts more than 500,000 landfills [1]. Landfills, once considered the final resting places for waste, are now being recognized as troves of valuable resources [2]. This paradigm shift has given rise to the resurgence of landfill mining—an increasingly popular practice. Landfill mining involves the extraction of valuable materials and resources from existing landfill sites [3], as well as the reduction in the environmental footprint of these sites [4,5]. Four categories of fractions can be obtained during landfill mining, namely, soil-like material or fine fraction; combustible fraction (including plastic, paper, wood, and textile); inert fraction (stone, glass, ceramic, and metal); and others consisting of the remaining fraction [3]. The fine fraction, the smallest fraction that is usually <10–25.4 mm depending on the study, stands out as a crucial yet often overlooked component, typically accounting for 40% to 70% of the total excavated materials [6–8]. Several studies have investigated the fine fraction's potential, highlighting its applicability in waste-to-energy conversion and the recovery of rare metals [9–13]. Extracting resources from this fraction is usually economically challenging, but this fraction has the potential for biocover construction on landfill sites [14].

Landfills are significant sources of greenhouse gas emissions, mainly methane. While methane collection systems are common in sanitary landfills, capturing high-quality gas with a substantial methane content is technically and economically infeasible in old landfills. Biocover emerges as a viable solution to mitigate low-level methane emissions. Methanotrophs play a crucial role in biocover functionality by utilizing methane as an energy source and microbially oxidizing it into carbon dioxide, thereby substantially reducing methane emissions [15]. This reduction aligns with the European Union's commitment to achieving climate neutrality by 2050 and supports EU decarbonization efforts toward the 2030 climate target plan and zero-pollution ambition [16].

One of the most critical parameters for biocover construction is the organic matter content, which is typically recommended to be more than 15%, according to Huber-Humer et al. [17]. While garden compost is commonly used for biocover construction [18–20], in this case, we consider alternative resources, as compost can be used more sustainably as a fertilizer. Consequently, searching for less valuable materials for biocover construction becomes crucial for sustainable waste management. As a potential biocover material, the fine fraction has received limited attention in previous studies, with practical investigations into its suitability for biocover construction conducted only in Sweden [11] and Estonia [16].

Traditionally, the research has focused on excavating and analyzing fine fraction landfill materials from a single depth or a limited range of depths within landfill sites [11,13,16]. While these studies have contributed valuable insights into these materials' physical and chemical properties, they often overlook the dynamic variations that occur at different depths within the landfill. Beyond examining fundamental parameters, this research incorporates an investigation of microplastics in evaluating the suitability of landfill fine fraction as a biocover material. Numerous studies have substantiated a significant abundance of microplastics within landfill bodies up to 83 particles/g [21–25] and their release with leachate [21,26,27]. Growing environmental concern about microplastic pollution relates to their pervasive presence, continual fragmentation, leaching of toxic additives, and integration into the trophic chain. Additionally, microplastics can absorb and transport organic pollutants, trace elements like heavy metals, and other detrimental agents, such as pharmaceuticals and pathogenic organisms, out of landfills [28].

The authors of this article hypothesized that the physical and chemical properties of fine fraction landfill materials vary significantly with depth within landfill sites, leading to the identification of depth-specific layers that may be more suitable for biocover construction. Additionally, considering previous research findings that indicate high microplastic abundance in landfills and a potential increase with depth [23], it is assumed that some layers, even if they exhibit the best suitability for biocovers, may present challenges due to the emission of microplastics. Therefore, this research has two-fold novelty: first, it seeks to analyze and compare fine fraction materials from varying depths within landfill sites, thereby providing a more holistic understanding of the dynamic properties of these materials throughout the landfill profile and choosing the best layer. Second, this study incorporates a rigorous investigation of microplastics as an integral part of the assessment. This inclusion is motivated by the urgent need to prevent microplastic leakage from biocovers, a concern that has been largely overlooked in previous research efforts.

So, the current article aims to analyze fine fraction landfill materials collected at different depths, focusing on their physical, chemical, and microplastic properties, in order to determine the optimal layer for extracting fine fractions and using them in biocover construction, contributing to sustainable waste management practices and enhancing control over landfill gas. Identifying the optimal layer holds significance for potential future landfill mining and reclamation operations, emphasizing the utilization of layers with the highest suitability, highest methane reduction potential, and minimal pollution levels.

2. Materials and Methods

2.1. Study Object

In the second stage of the project, titled "Assessment and Testing of Resource Recovery from Landfills in Lithuanian Conditions", drilling operations were conducted on 28 April 2022 at the closed Kuršėnai landfill, located in the Šiauliai waste management region, with the coordinates of 55°55′05.4″ N 22°52′55.8″ E (Figure 1). The objective of the drilling was to analyze the morphological composition of the landfilled waste and assess the feasibility of landfill mining operations. The Kuršėnai landfill was operational from 1970 to 2007. No biogas collection system is in place, and methane emissions remain high despite landfill age. Given these circumstances, a decision was made to investigate the suitability of the fine fractions of the landfill for the construction of a biocover.

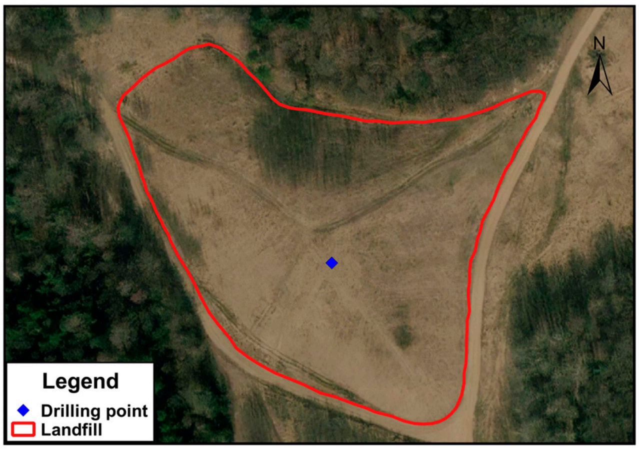

Figure 1. Kuršėnai landfill and drilling point.

Rotary-spoke drilling was carried out using the Wamet MWG-6 (small-size caterpillar drilling rig) at the center of the landfill. Each drill had a length of 1.5 m, and since the drilling was conducted to a depth of 10.5 m, a total of 7 drills were used. Initially, the cover layer was removed, and then every 1.5 m, the entire mass drilled from that interval, typically 10–15 kg as a mixed sample, was collected. Subsequently, this mass was sieved through a 20 mm sieve to separate the fine fractions. The coarse fractions were further sorted into different fractions for morphological analysis. Three replicant samples of fine fractions weighing 1 kg were then collected, transported to the laboratory, and subjected to further analysis. On average, the fine fractions accounted for approximately 74.6% of the total material collected.

2.2. Physical and Chemical Characterization

In this study, the fine fractions from landfill sites were characterized following the methodology outlined by Huber-Humer et al. [17] for assessing their suitability as a methane (CH_4) oxidation substrate. Huber-Humer et al. [17] developed the concepts of biocover and target values for biocover material, which are used as references in many studies [11,16,29]. The characterization process encompassed an array of key parameters, including organic matter (OM) content, moisture content (MC), water-holding capacity (WHC), particle size distribution (PSD), bulk density (BD), pH, and the electrical conductivity (EC) of leachate. In addition to the basic parameters, heavy metals and numerical microplastic concentrations were analyzed.

MC was determined by drying at 105 °C for 24 h and OM by loss on ignition at 550 °C for 2 h. The WHC was calculated using the gravimetric method as the weight difference before and after saturation with water. The bulk density was measured according to ISO 11885. PSD was analyzed in two options: first, following Huber-Humer's recommendation of 6–20 mm, 2–6 mm, and <2 mm; for the second option, categories were chosen as

10–20 mm, 5–10 mm, 2–5 mm, 1–2 mm, 0.5–1 mm, 0.25–0.5 mm, 0.125–0.25 mm, and <0.125 mm.

The electrical conductivity and pH were measured with pH/Cond340i (WTW) and PL-700PVS meters at a dilution ratio of 1:10 (10 g of fine fraction was mixed with 100 mL of distilled water) according to Pansu and Gautheyrou [30] and Kriipsalu [16].

Heavy metals underwent analysis using inductively coupled plasma optical emission spectroscopy (ICP-OES). Fine fraction samples (0.32 g–0.52 g) were mineralized with 6 mL of concentrated nitric acid and 2 mL of hydrofluoric acid, hydrochloric acid, and hydrogen peroxide at 1200 W, 6 MPa, and pRate: 30 kPa·s^{-1} (Multiwalve 3000). After the mineralization, the solution was poured into 50 mL flasks and diluted to 40 mL using deionized water. The analysis of the solutions (As, Cd, Cr, Ni, Pb, Zn, Cu, and Fe) was performed using an ICP-OES, Optima 8000 (Perkin Elmer). The scanning of each single sample during element analysis was repeated three times to gather reasonably good results. An analysis was conducted in two replicates of each sample. The mercury concentration was determined according to [31,32].

2.3. Concentration of Microplastics

Fine fractions < 20 mm were dried at 80 °C for 24 h to assess the microplastic concentration at each depth. Then, three 10 g samples per depth were sieved through 1 mm; in this article, a concentration of microplastics of 50–1000 µm was analyzed. Microplastic extraction and identification were described in detail in previous articles [23,33]. Microplastics < 50 µm were not studied due to the limitations of the chosen identification method. In short, organic matter was removed using Fenton's reagent with heating. Subsequently, inorganic particles were separated using density separation with potassium formate and vacuum-filtrated on glass fiber filters. Then, microplastic particles were identified by staining the filters with Nile red dye, and they were examined under a fluorescence microscope on a blue excitation scale. The quantity and size of microplastics were analyzed in ImageJ 1.52v software.

2.4. Data Analysis

All data are presented as means ± SD (standard deviation). The normality of the data was assessed through a combination of visual observations using histograms and a statistical test, specifically the Shapiro–Wilk test ($p < 0.05$). All datasets deviated from a normal distribution; therefore, the Spearman correlation analysis was subsequently employed to explore relationships between various variables.

3. Results and Discussion

3.1. Physical Parameters

Physical parameters of fine fractions from different depths are presented in Figures 2 and 3. Moisture content varied from 20.99 ± 1.14% at a depth of 9–10.5 m to 36.01 ± 0.31% at a depth of 0–1.5 m. According to Huber-Hummer et al. [17], the ideal range of moisture for biocover material should be 30–50%; therefore, except for samples from the deepest depth, all samples met the recommended moisture content. The biogas flowing through the biocover may experience heating due to elevated temperatures, leading to water evaporation. Despite the water produced during the methane oxidation process, there is a possibility of desiccation in the biocover and a decrease in the efficiency of methane reduction [34]. Methanotrophic microorganisms tend to become inactive enzymes at less than 13% water content [17]. All samples demonstrated sufficient WHC in terms of recommended values (50–120%). The water-holding capacity was lowest at a depth of 6–7.5 m, measuring 73.8 ± 3.88%, while it was the highest at a depth of 4.5–6 m. The bulk density ranged from 0.71 ± 0.02 kg/L to 1.01 ± 0.03 kg/L. In samples from a Swedish landfill [11], the BD was lower (0.69 kg/L) than found in this article and lower than the recommended value of 0.8–1.1 kg/L. In our study, the BD in only two layers was lower than the recommended value, highlighting the importance of an in-depth analysis of fine fraction parameters. Bulk density is a critical parameter for efficient biocover functionalization, as it influences porosity and the movement of air and gases, particularly oxygen and methane [35]. Adequate aeration supports the activity of aerobic microorganisms involved in methane oxidation and impacts the efficiency of the reduction in methane emissions.

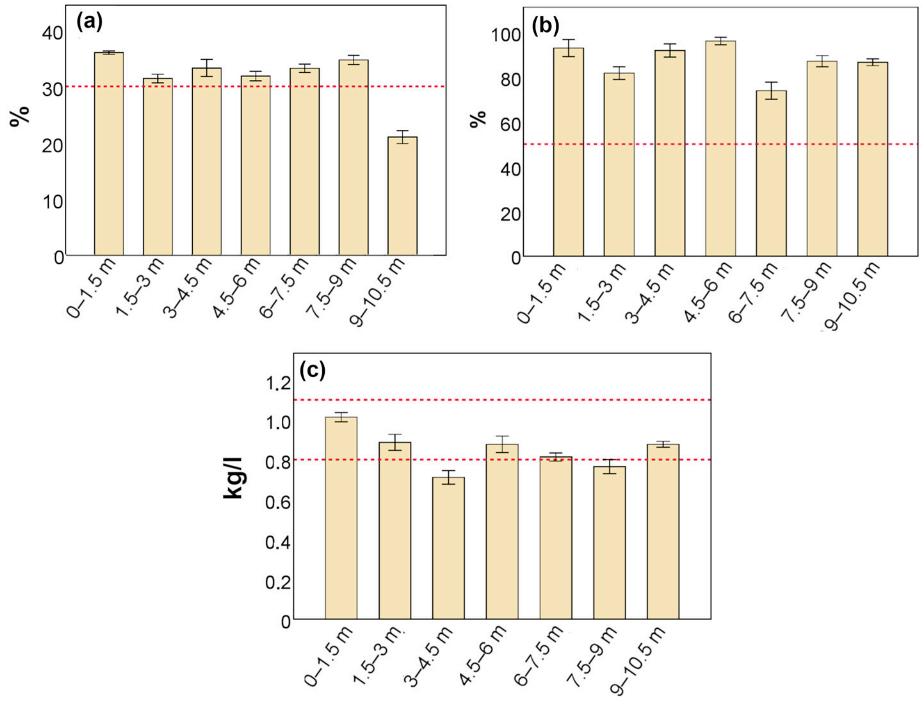

Figure 2. Physical parameters of fine fraction: (**a**) moisture content; (**b**) water-holding capacity; and (**c**) bulk density. Red dotted lines—recommended values. Error bars ± 1 SD.

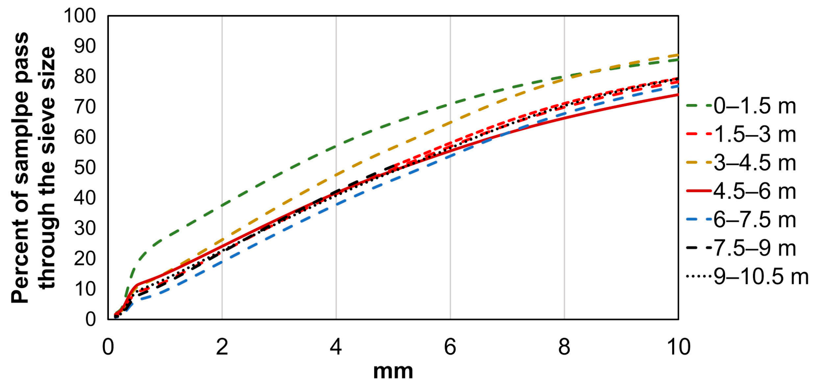

Figure 3. Particle size distribution of fine fractions from different depths.

Particle size distribution is presented in Figure 3 and, according to Huber-Humer et al. [17], in Table S1. For well-balanced materials suitable for biocover applications, it is recommended that approximately 20–30% of the material pass through a 2 mm sieve, roughly 40% pass through a 2–6.3 mm sieve, about 20–40% pass through a 6.3–20 mm sieve, and the remaining 10% consists of particles larger than 20 mm.

In our case, we tested the fine fraction (particles < 20 mm), and the results indicate that all samples exhibit a normal particle size distribution, aligning well with the recommended criteria for biocover materials. However, it is noteworthy that samples from depths of 0–1.5 m exhibited a higher percentage of material passing through the 2 mm sieve, measuring 38.39%. This percentage is slightly above the recommended range and balance for ideal biocover material, which can impact porosity and gas diffusion in the biocover layer. Conversely, samples from depths of 6–7.5 m exhibited a slightly lower percentage, at 16.18%, which falls below the recommended range for the 2 mm sieve fraction.

3.2. Chemical Parameters

Figure 4 presents the OM of fine fractions at different depths. The OM content varied from 13.75 ± 0.5% at a depth of 9–10.5 m to 18.55 ± 0.59% at a depth of 7.5–9 m. As in our study, an old landfill was investigated, and such a value of OM is usual. Mönkäre et al. [13] analyzed fine fractions from two landfills in Finland with ages ranging from 1 to 10 years and 24 to 40 years and found a higher content of OM in the young landfill compared to the old one—15.5–27.3% and 6.0–24.0%, respectively. Our analysis also demonstrates that organic content in samples from the 0–1.5, 3–4.5, 4.5–6, and 7.5–9 m depths met the 15% recommended value, while other depths were characterized with a slightly lower content. The lower level of organic content leads to a reduction in methane removal efficiency. Organic matter is a nutrient source for the microorganisms involved in methane oxidation and is needed for their growth and activity [36,37].

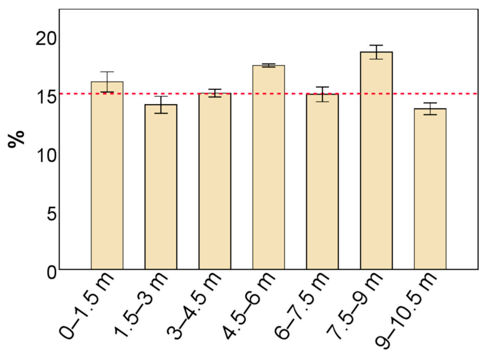

Figure 4. Organic matter content. Red dotted line—recommended value. Error bars ± 1 SD.

pH and conductivity are other important parameters that directly impact the vitality of methanotrophic microorganisms. The pH of leachate varied from 7.99 to 8.44 and was alkaline, which is a common finding and is connected to the age of the landfill (Figure 5) [38]. All samples fall within the recommended pH range for methanotrophs' activity, ranging from 6.5 to 8.5, but for some samples, the pH was close to the upper limit of 8.5.

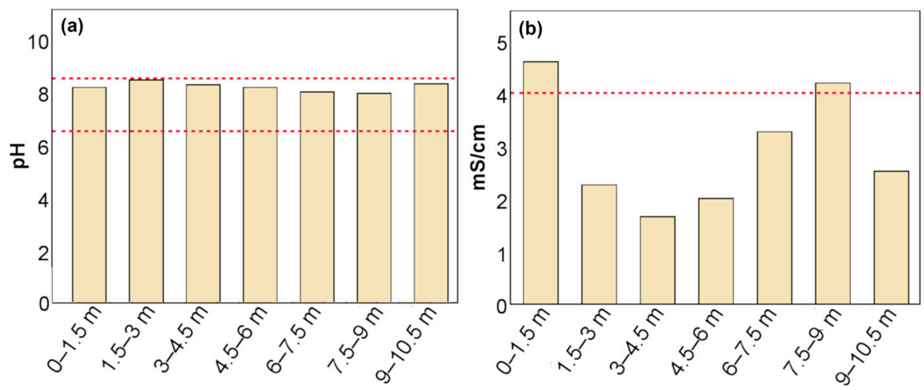

Figure 5. pH (**a**) and conductivity (**b**) of leachate. Red dotted lines—recommended values.

The conductivity values show a significant variation ranging from 1660 µS/cm at a depth of 3–4.5 m to 4592 µS/cm at a depth of 0–1.5 m. According to Huber-Humer et al. [17], the conductivity value for biocover should be no more than 4000 µS/cm. In depths 0–1.5 and 7.5–9 m, the conductivity was slightly higher—4592 µS/cm and 4188 µS/cm, respectively. As methanotrophs are rather tolerant to higher conductivity [17], using fine fractions from these depths as the main material for biocover is limited.

The average concentration of heavy metals at all depths is provided in Table 1. These concentrations were compared with the limit values established by Lithuanian legislation [39]. The table includes the limit values for both industrial soils and forested areas, considering the potential change in land use after biocover construction and remediation.

Table 1. Average concentration of heavy metals in different landfill depths, mg/kg.

	Depths							Soil for Industry Zone	Soil for Forest Areas
	0–1.5 m	**1.5–3 m**	**3–4.5 m**	**4.5–6 m**	**6–7.5 m**	**7.5–9 m**	**9–10.5 m**		
As				<1.35				80	30
Cd				<0.51				120	60
Cr	67.27	37.25	48.98	51.25	44.12	68.39	63.59	600	300
Cu	209.22	89.86	74.16	123.99	91.84	136.30	210.19	200	100
Ni	4.57				<0.42			300	150
Pb	31.63	5.24	<1.20	91.99	<1.20	105.33	88.37	500	150
Zn	281.02	465.09	217.64	367.20	262.19	589.16	244.35	1200	600
Hg	<0.02	0.035	0.11	0.06	0.10	<0.02	<0.02	1.00	0.75
Fe	10,817.1	7517.5	10,665.6	8566.9	7470.1	11,042.7	10,338.2		

From Table 1, it is evident that the concentration of heavy metals was low and was found to be below the regulatory limits set by Lithuanian regulation for both industrial soils and forested areas, except for the concentration of Cu in the top and bottom layers. In addition, in the layer of 7.5–9 m, the concentration of Zn and Pb was the highest among other depths and close to the limit for soil in forested areas. However, heavy metals, with their low concentrations, are well sorbed and do not leak out, as was found in studies by Kaczala et al. [40] and Burlakovs et al. [9]. It should be noted that there does not appear to be a consistent trend in concentration change with depth, likely because the concentration is highly dependent on the composition of waste deposited at a particular time.

3.3. Concentration of Microplastics and Size Distribution

Landfill mining involves excavating and disturbing the waste, which could release microplastics into the environment. These microplastics could then contaminate surrounding ecosystems and bodies of water, and they can even enter the food chain, affecting both wildlife and human health.

Numerical concentrations of microplastics are presented in Figure 6. Our study revealed a significant and concerningly high concentration of microplastics in the fine fraction of the landfill, making its use for remediation challenging. The average concentration of microplastics ranges from 4958 ± 439 particles/kg at a 0–1.5 m depth to 34,917 ± 1922 particles/kg at a 6–7.5 m depth. The abundance of microplastics in soil [41,42], green and food compost [43], and stabilized organic output from MBT [44] is usually lower than the abundance found in landfill bodies.

It was noted that the concentration of microplastics increases up to a depth of 6–7.5 m, and this rise can be attributed to the migration of microplastics with leachate from the upper layers that are deeper into the landfill profile. As a result, this migration and redistribution of microplastics cause their accumulation at intermediate depths. However, beyond the depth of 6–7.5 m, a decrease in the concentration of microplastics is observed. Several factors can contribute to this decline. Firstly, the morphological analysis shows that the concentration of plastic content (Table S2) in the landfill waste also increases and then decreases as we move deeper into the landfill. Correlation analysis shows a medium–high positive relationship between the concentration of microplastics and plastic content (r = 0.757). Secondly, the landfill body is not homogenous, and localized barriers may form within the waste profile. These barriers can obstruct the further migration of microplastics, causing them to accumulate at certain depths.

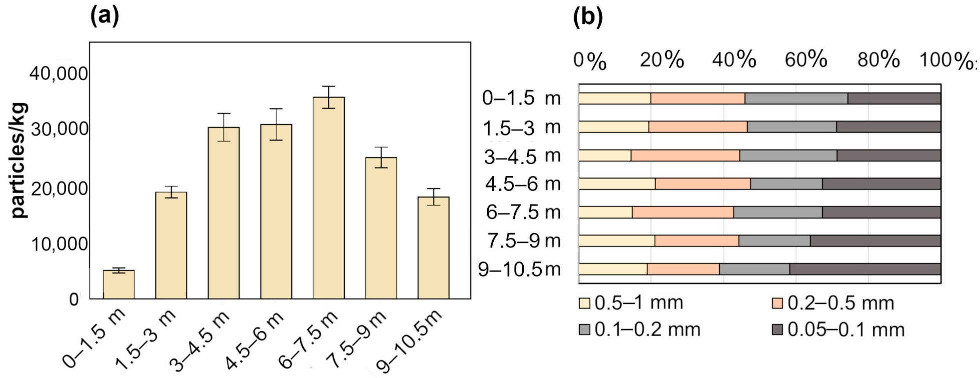

Figure 6. (**a**) Numerical concentration of microplastics. Error bars ± 1 SD; (**b**) size distribution of identified microplastics.

Consequently, the accumulation of microplastics in specific locations due to these barriers could result in decreased concentrations in other sections of the landfill. Moreover, microplastic degradation might occur more rapidly under landfill conditions. This degradation process could contribute to the overall reduction in the concentration of microplastics in landfills. However, the faster degradation alone does not fully explain the sharp decrease observed after the initial increase.

The size distribution of microplastics presented in Figure 6b reveals an intriguing trend: the percentage of the finest microplastics, measuring between 0.05 and 0.1 mm, appears to increase with depth. This phenomenon could be attributed to the degradation of plastic materials over time. However, it is essential to underscore that finer microplastics pose a more significant threat to the environment due to their ease of entry into our trophic chain. Consequently, when selecting the depth for fine fraction extraction and its subsequent use in biocover construction, special attention should be devoted to these finer microplastics to ensure the overall effectiveness of environmental management strategies. Therefore, the first two layers, in the context of the concentration of microplastics and their size distribution, have a lesser environmental impact during biocover construction.

A Spearman correlation analysis was also conducted to examine the relationship between microplastics and heavy metal concentrations. The results revealed a strong positive correlation with mercury (Hg) concentration ($r = 0.82$, $p < 0.05$). Previous studies revealed that microplastics can absorb heavy metals and become more toxic [45,46]. Therefore, opting for a layer with a high concentration of microplastics for biocover construction entails a dual risk, as microplastics can adsorb and transport heavy metals and other pollutants.

3.4. Comprehensive Assessment of Fine Fraction Suitability for Biocover

The utilization of fine fraction landfill materials for biocover construction serves as an exemplary embodiment of circular economy and sustainable waste management. Circular economy emphasizes the reduction in waste, the efficient use of resources, and the repurposing of materials to create a closed-loop system [47]. In this context, repurposing fine fraction materials from landfills aligns perfectly with these principles. By transforming a previously discarded waste fraction into a valuable resource for mitigating environmental impacts, such as landfill gas emissions, we embrace a circular approach.

Samples of the fine fraction collected from seven different depths were analyzed for several key parameters to assess their suitability as biocover construction materials. The comprehensive results of this analysis are presented in Table 2. In terms of factors such as moisture content and water-holding capacity, samples from all depths met the requirements. However, it is noteworthy that the organic matter content, one of the most important parameters, only met the criteria in four layers.

Table 2. Summary table of values for various parameters of the fine fractions (bold—under/above the recommended values).

	Parameters							
	pH	**Conductivity, mS/cm**	**Organic Content, %**	**Moisture Content, %**	**Bulk Density, kg/l**	**WHC, %**	**Microplastics, Particle/kg**	**Heavy Metals, mg/kg**
target value	6.5–8.5	4000	15	30–50	0.8–1.1	50–130	as low as possible	separate per each metal
0–1.5 m	8.16	**4592**	16.02	36.01	1.01	92.9	4958	Cu exceeds the limit
1.5–3 m	8.44	2260	**14.06**	31.41	0.89	81.6	18,500	below the limit
3–4.5 m	8.26	1660	15.05	33.27	**0.71**	91.8	29,708	
4.5–6 m	8.16	2000	17.42	31.82	0.88	96	30,208	
6–7.5 m	7.99	3265	**14.95**	33.22	0.81	73.8	34,917	
7.5–9 m	7.93	**4188**	18.55	34.70	**0.76**	87	24,500	
9–10.5 m	8.29	2520	**13.75**	**20.99**	0.87	86.6	17,667	Cu exceeds the limit

Considering all these parameters, it becomes evident that the layer at a depth of 4.5–6 m appears to be the most favorable choice for biocover construction. This depth range exhibits a promising combination of suitable characteristics. However, it is worth noting that the concentration of microplastics in this layer is relatively high.

On the other hand, the top layer also demonstrates good suitability for biocover construction, maintaining a lower concentration of microplastics. However, the conductivity is the highest, and the concentration of copper exceeds the limits. As microplastics are intolerant of high conductivity, the use of this layer for biocover construction is limited. The layer of 1.5–3 m is also suitable according to all parameters except for organic matter content. It is essential to highlight that parameter values can be managed and controlled via a strategic mixing of the fine fractions with appropriate amendments. Therefore, the suitability of the layer of 1.5–3 m can be increased by mixing the fine fraction with sewage sludge or green compost to decrease the concentration of microplastics. In a relevant study, Kriipsalu et al. [16] examined fine fraction materials from an Estonian landfill, and, to increase organic content, the fine fraction was mixed with sewage sludge compost in a proportion of 3:1.

The layer of 3–4.5 m has low bulk density, which can be increased by mixing it with soil, sand, or bottom ash. However, this layer was also characterized by a relatively high concentration of microplastics. The layer of 6–7.5 m had the highest concentration of microplastics and, therefore, is not recommended for use as a material for biocover construction. The layer of 7.5–9 m had low bulk density and high conductivity, so its use for biocover is limited due to the intolerance of methanotrophs. The last layer had a low organic and moisture content, but it can be increased by mixing it with compost or sludge. A correlation analysis revealed several medium–high (0.5–0.8) relationships between tested parameters, such as the relationship between the pH and OM content (r = −0.74), pH and MC (r = −0.65), OM and MC (r = 0.71), pH and conductivity (r = 0.52), WHC and OM (r = 0.64), and bulk density and microplastics (r = −0.61); however, only the first two were statistically significant ($p < 0.05$) (Table 3).

Table 3. Correlation coefficients (r) between analyzed parameters.

	pH	Conductivity	Organic Content	Moisture Content	Bulk Density	WHC	Microplastics
pH		**0.52, $p > 0.05$**	**−0.74, $p < 0.05$**	**−0.65, $p > 0.05$**	0.32	−0.13	−0.39
Conductivity	**0.52, $p > 0.05$**		0.21	0.5	0.32	−0.14	−0.45
Organic content	**−0.74, $p < 0.05$**	0.21		**0.71, $p < 0.05$**	−0.14	**0.64, $p > 0.05$**	0.18
Moisture content	**−0.65, $p > 0.05$**	0.5	**0.71, $p < 0.05$**		−0.11	0.39	−0.04
Bulk density	0.32	0.32	−0.14	−0.11		0.18	**−0.61, $p > 0.05$**
WHC	−0.13	−0.14	**0.64, $p > 0.05$**	0.39	0.18		−0.18
Microplastics	−0.39	−0.45	0.18	−0.04	**−0.61, $p > 0.05$**	−0.18	

The positive correlation between moisture and organic matter content is often observed among other studies [10]. Organic matter, such as food waste, paper, and plant materials, can be hygroscopic, meaning it has an inherent ability to absorb and retain moisture from the surrounding environment. This fact can also explain the positive correlation between OM and WHC. Also, some organic waste categories possess a higher moisture content and, during decomposition, can produce leachate [48].

A negative correlation between pH and organic content, as well as pH and moisture, can be connected to leachate formation. As organic matter decomposes, it releases moisture into the landfill, creating leachate. Leachate can have a lower pH due to organic acids forming during decomposition [49]. A negative correlation between OM and pH was also confirmed by Lindamulla et al. [50]. The correlation between the concentration of microplastics and bulk density is not statistically significant, and there is no explanation for this in the literature. However, according to the authors' presumption, lower bulk density, associated with higher porosity, provides more pore space availability for microplastics to occupy, leading to a negative correlation. Additionally, lower bulk density may facilitate better water infiltration, potentially carrying microplastics downward and resulting in their accumulation in lower layers.

4. Conclusions

The fine fraction from the diverse depths of the Kursenia landfill underwent an extensive assessment across various parameters to ascertain its aptness as a construction material for a biocover. The findings delineated substantial variations in the characteristics of fine fractions at different depths. After an extensive analysis of all key parameters, it was found that some depths are more suitable for fine fraction extraction. In our study, the fine fraction from the 4.5–6 m depth at the Kuršėnai landfill emerged as the most suitable material for biocover construction based on its physical and chemical attributes. However, this layer revealed the second-highest concentration of microplastics, totaling 30,208 ± 273 particles/kg. This significant microplastic content poses a substantial challenge, restricting immediate application and necessitating solutions to enhance other fractions with lower concentrations of microplastics. Elevated concentrations of microplastics raise environmental concerns, particularly during landfill mining operations, which may release microplastics into the ecosystem.

Considering both material properties and pollution concentrations, the layer of 1.5–3 emerges as a preferable option for biocover construction, meeting essential requirements, except for organic content, and demonstrating significantly lower microplastic levels. The shortfall in organic content can be addressed by blending it with organic-rich materials like green compost or sludge.

Within this research, several moderate correlations between parameters were observed, including the crucial relationship between microplastics and mercury concentration (r = 0.82). These findings underscore the dual risk associated with microplastic pollution, emphasizing its capacity to absorb and transport heavy metals, increasing overall toxicity.

Our study underscores the vital importance of extensive investigations into material parameters and pollution concentrations before undertaking landfill mining and biocover construction. This meticulous analysis is essential for selecting the most effective layer for methane degradation while mitigating additional environmental impact.

The utilization of fine fraction as a biocover material is a notable example of a circular economy, repurposing waste fractions and minimizing landfill methane emissions with minimal reliance on additional materials. Moreover, this aligns with the European Union's commitment to reduce greenhouse gas emissions. However, there remain unexplored aspects, and future research is warranted to delve into areas such as the release of microplastics from biocover, monitoring the efficiency of biocovers made of fine fractions on actual landfills, and enhancing the efficiency of fine fractions in methane oxidation.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/su152416914/s1, Table S1: Particle size distribution; Table S2: Plastic content in different depths according to morphological analysis.

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References

1. EUROLCO Data Launched on the Landfill Situation in the EU-28. Available online: https://eurelco.org/2018/09/30/data-launched-on-the-landfill-situation-in-the-eu-28/ (accessed on 30 September 2023).
2. Krook, J.; Svensson, N.; Eklund, M. Landfill Mining: A Critical Review of Two Decades of Research. *Waste Manag.* **2012**, *32*, 513–520. [CrossRef]
3. Jain, M.; Kumar, A.; Kumar, A. Landfill Mining: A Review on Material Recovery and Its Utilization Challenges. *Process Saf. Environ. Prot.* **2023**, *169*, 948–958. [CrossRef]
4. Joseph, K.; Nagendran, R.; Palanivelu, K.; Thanasekaran, K.; Visvanathan, C. *Dumpsite Rehabilitation and Landfill Mining. ARRPER Report*; Anna University: Chennai, India, 2004; pp. 1–53.
5. Burlakovs, J.; Kriipsalu, M.; Klavins, M.; Bhatnagar, A.; Vincevica-Gaile, Z.; Stenis, J.; Jani, Y.; Mykhaylenko, V.; Denafas, G.; Turkadze, T.; et al. Paradigms on Landfill Mining: From Dump Site Scavenging to Ecosystem Services Revitalization. *Resour. Conserv. Recycl.* **2017**, *123*, 73–84. [CrossRef]
6. Hernández Parrodi, J.C.; Höllen, D.; Pomberger, R. Characterization of Fine Fractions from Landfill Mining: A Review of Previous Investigations. *Detritus* **2018**, *2*, 46. [CrossRef]
7. Hull, R.M.; Krogmann, U.; Strom, P.F. Composition and Characteristics of Excavated Materials from a New Jersey Landfill. *J. Environ. Eng.* **2005**, *131*, 478–490. [CrossRef]
8. Quaghebeur, M.; Laenen, B.; Geysen, D.; Nielsen, P.; Pontikes, Y.; Van Gerven, T.; Spooren, J. Characterization of Landfilled Materials: Screening of the Enhanced Landfill Mining Potential. *J. Clean. Prod.* **2013**, *55*, 72–83. [CrossRef]
9. Burlakovs, J.; Jani, Y.; Kriipsalu, M.; Vincevica-Gaile, Z.; Kaczala, F.; Celma, G.; Ozola, R.; Rozina, L.; Rudovica, V.; Hogland, M.; et al. On the Way to 'Zero Waste' Management: Recovery Potential of Elements, Including Rare Earth Elements, from Fine Fraction of Waste. *J. Clean. Prod.* **2018**, *186*, 81–90. [CrossRef]
10. Chandana, N.; Goli, V.S.N.S.; Mohammad, A.; Singh, D.N. Characterization and Utilization of Landfill-Mined-Soil-Like-Fractions (LFMSF) for Sustainable Development: A Critical Appraisal. *Waste Biomass Valorization* **2021**, *12*, 641–662. [CrossRef]

11. Jani, Y.; Kaczala, F.; Marchand, C.; Hogland, M.; Kriipsalu, M.; Hogland, W.; Kihl, A. Characterisation of Excavated Fine Fraction and Waste Composition from a Swedish Landfill. *Waste Manag. Res. J. Sustain. Circ. Econ.* **2016**, *34*, 1292–1299. [CrossRef]
12. Parrodi, J.C.H.; Höllen, D.; Pomberger, R. Potential and Main Technological Challenges for Material and Energy Recovery from Fine Fractions of Landfill Mining: A Critical Review. *Detritus* **2018**, *3*, 19–29. [CrossRef]
13. Mönkäre, T.J.; Palmroth, M.R.T.; Rintala, J.A. Characterization of Fine Fraction Mined from Two Finnish Landfills. *Waste Manag.* **2016**, *47*, 34–39. [CrossRef]
14. Pehme, K.M.; Orupõld, K.; Kuusemets, V.; Tamm, O.; Jani, Y.; Tamm, T.; Kriipsalu, M. Field Study on the Efficiency of a Methane Degradation Layer Composed of Fine Fraction Soil from Landfill Mining. *Sustainability* **2020**, *12*, 6209. [CrossRef]
15. Scheutz, C.; Duan, Z.; Møller, J.; Kjeldsen, P. Environmental Assessment of Landfill Gas Mitigation Using Biocover and Gas Collection with Energy Utilisation at Aging Landfills. *Waste Manag.* **2023**, *165*, 40–50. [CrossRef]
16. Kriipsalu, M.; Somani, M.; Pehme, K.; Tamm, O.; Truu, J.; Truu, M.; Orupold, K. Performance of Biocover in Controlling Methane Emissions from Landfill: A Decade of Full-Scale Investigation. *Process Saf. Environ. Prot.* **2023**, *172*, 486–495. [CrossRef]
17. Huber-Humer, M.; Röder, S.; Lechner, P. Approaches to Assess Biocover Performance on Landfills. *Waste Manag.* **2009**, *29*, 2092–2104. [CrossRef] [PubMed]
18. Niemczyk, M.; Berenjkar, P.; Wilkinson, N.; Lozecznik, S.; Sparling, R.; Yuan, Q. Enhancement of CH4 Oxidation Potential in Bio-Based Landfill Cover Materials. *Process Saf. Environ. Prot.* **2021**, *146*, 943–951. [CrossRef]
19. Pedersen, G.B.; Scheutz, C.; Kjeldsen, P. Availability and Properties of Materials for the Fakse Landfill Biocover. *Waste Manag.* **2011**, *31*, 884–894. [CrossRef]
20. Scheutz, C.; Pedersen, R.B.; Petersen, P.H.; Jørgensen, J.H.B.; Ucendo, I.M.B.; Mønster, J.G.; Samuelsson, J.; Kjeldsen, P. Mitigation of Methane Emission from an Old Unlined Landfill in Klintholm, Denmark Using a Passive Biocover System. *Waste Manag.* **2014**, *34*, 1179–1190. [CrossRef]
21. Su, Y.; Zhang, Z.; Wu, D.; Zhan, L.; Shi, H.; Xie, B. Occurrence of Microplastics in Landfill Systems and Their Fate with Landfill Age. *Water Res.* **2019**, *164*, 114968. [CrossRef]
22. Puthcharoen, A.; Leungprasert, S. Determination of Microplastics in Soil and Leachate from the Landfills. *Thai Environ. Eng. J.* **2019**, *33*, 39–46.
23. Sholokhova, A.; Denafas, G.; Ceponkus, J.; Omelianenko, T. Microplastics in Landfill Bodies: Abundance, Spatial Distribution and Effect of Landfill Age. *Sustainability* **2023**, *15*, 5017. [CrossRef]
24. Goli, V.S.N.S.; Singh, D.N. Extraction and Characterization of Microplastics in Landfill-Mined-Soil-like-Fractions: A Novel Methodology. *Chem. Eng. J.* **2023**, *452*, 139217. [CrossRef]
25. Sima, J.; Wang, J.; Song, J.; Du, X.; Lou, F.; Pan, Y.; Huang, Q.; Lin, C.; Wang, Q.; Zhao, G. Dielectric Barrier Discharge Plasma for the Remediation of Microplastic-Contaminated Soil from Landfill. *Chemosphere* **2023**, *317*, 137815. [CrossRef] [PubMed]
26. He, P.; Chen, L.; Shao, L.; Zhang, H.; Lü, F. Municipal Solid Waste (MSW) Landfill: A Source of Microplastics? -Evidence of Microplastics in Landfill Leachate. *Water Res.* **2019**, *159*, 38–45. [CrossRef] [PubMed]
27. van Praagh, M.; Hartman, C.; Brandmyr, E. *Microplastics in Landfill Leachates in the Nordic Countries*; Nordic Council of Ministers: Copenhagen, Denmark, 2019. [CrossRef]
28. Wojnowska-Baryła, I.; Bernat, K.; Zaborowska, M. Plastic Waste Degradation in Landfill Conditions: The Problem with Microplastics, and Their Direct and Indirect Environmental Effects. *Int. J. Environ. Res. Public Health* **2022**, *19*, 13223. [CrossRef] [PubMed]
29. Bajwa, T.M.; Fall, M.; Alshawmar, F. Experimental Characterization of the Engineering Properties of Landfill Compost-Biocover. *Appl. Sci.* **2022**, *12*, 4276. [CrossRef]
30. Pansu, M.; Gautheyrou, J. *Handbook of Soil Analysis: Mineralogical, Organic and Inorganic Methods*; Springer: Berlin/Heidelberg, Germany, 2006; pp. 1–993. [CrossRef]
31. *ISO 16968:2015*; Solid Biofuels—Determination of Minor Elements. International Organization for Standardization: Geneva, Switzerland, 2015.
32. *ISO 12846:2012*; Water Quality—Determination of Mercury. International Organization for Standardization: Geneva, Switzerland, 2012.
33. Sholokhova, A.; Ceponkus, J.; Sablinskas, V.; Denafas, G. Abundance and Characteristics of Microplastics in Treated Organic Wastes of Kaunas and Alytus Regional Waste Management Centres, Lithuania. *Environ. Sci. Pollut. Res. Int.* **2022**, *29*, 20665–20674. [CrossRef]
34. Cossu, R.; Stegmann, R. *Solid Waste Landfilling: Concepts, Processes, Technologies*; Elsevier: Amsterdam, The Netherlands, 2018; ISBN 0128183365.
35. Gebert, J.; Huber-Humer, M.; Cabral, A.R. Design of Microbial Methane Oxidation Systems for Landfills. *Front. Environ. Sci.* **2022**, *10*, 907562. [CrossRef]
36. Hrad, M. *Quantification of Landfill Gas Emissions in Biocovers—An Experimental Simulation in Lysimeters*; Universitätsbibliothek Bodenkultur: Wien, Austria, 2010.
37. Pariatamby, A.; Cheah, W.Y.; Shrizal, R.; Thamlarson, N.; Lim, B.T.; Barasarathi, J. Enhancement of Landfill Methane Oxidation Using Different Types of Organic Wastes. *Environ. Earth Sci.* **2015**, *73*, 2489–2496. [CrossRef]
38. Singh, A.; Chandel, M.K. Physicochemical and FTIR Spectroscopic Analysis of Fine Fraction from a Municipal Solid Waste Dumpsite for Potential Reclamation of Materials. *Waste Manag. Res. J. Sustain. Circ. Econ.* **2020**, *39*, 374–385. [CrossRef]

39. D1-230 Dėl Cheminėmis Medžiagomis Užterštų Teritorijų Tvarkymo Aplinkos/Apsaugos Reikalavimų Patvirtinimo. Available online: https://e-seimas.lrs.lt/portal/legalAct/lt/TAD/TAIS.319604/aWhXQOotGB (accessed on 11 October 2023).
40. Kaczala, F.; Orupõld, K.; Augustsson, A.; Burlakovs, J.; Hogland, M.; Bhatnagar, A.; Hogland, W. Fractionation of Pb and Cu in the Fine Fraction (<10 Mm) of Waste Excavated from a Municipal Landfill. *Waste Manag. Res.* **2017**, *35*, 1175–1182. [CrossRef]
41. Büks, F.; Kaupenjohann, M. Global Concentrations of Microplastics in Soils—A Review. *SOIL* **2020**, *6*, 649–662. [CrossRef]
42. Medynska, A.; Medynska-Juraszek, M.; Szczepá, A. Microplastic Pollution in EU Farmland Soils: Preliminary Findings from Agricultural Soils (Southwestern Poland). *Agriculture* **2023**, *13*, 1733. [CrossRef]
43. Porterfield, K.K.; Hobson, S.A.; Neher, D.A.; Niles, M.T.; Roy, E.D. Microplastics in Composts, Digestates, and Food Wastes: A Review. *J. Environ. Qual.* **2023**, *52*, 225–240. [CrossRef]
44. Sholokhova, A.; Denafas, G.; Mykhaylenko, V. Microplastics Generation and Concentration during Mechanical-Biological Treatment of Mixed Municipal Solid Waste. *Environ. Res.* **2022**, *214*, 113815. [CrossRef]
45. Khalid, N.; Aqeel, M.; Noman, A.; Khan, S.M.; Akhter, N. Interactions and Effects of Microplastics with Heavy Metals in Aquatic and Terrestrial Environments. *Environ. Pollut.* **2021**, *290*, 118104. [CrossRef]
46. Liu, Q.; Wu, H.; Chen, J.; Guo, B.; Zhao, X.; Lin, H.; Li, W.; Zhao, X.; Lv, S.; Huang, C. Adsorption Mechanism of Trace Heavy Metals on Microplastics and Simulating Their Effect on Microalgae in River. *Environ. Res.* **2022**, *214*, 113777. [CrossRef]
47. Korhonen, J.; Honkasalo, A.; Seppälä, J. Circular Economy: The Concept and Its Limitations. *Ecol. Econ.* **2018**, *143*, 37–46. [CrossRef]
48. Jain, M.S.; Daga, M.; Kalamdhad, A.S. Variation in the Key Indicators during Composting of Municipal Solid Organic Wastes. *Sustain. Environ. Res.* **2019**, *29*, 9. [CrossRef]
49. Abdel-Shafy, H.I.; Ibrahim, A.M.; Al-Sulaiman, A.M.; Okasha, R.A. Landfill Leachate: Sources, Nature, Organic Composition, and Treatment: An Environmental Overview. *Ain Shams Eng. J.* **2023**, *15*, 102293. [CrossRef]
50. Lindamulla, L.; Nanayakkara, N.; Othman, M.; Jinadasa, S.; Herath, G.; Jegatheesan, V. Municipal Solid Waste Landfill Leachate Characteristics and Their Treatment Options in Tropical Countries. *Curr. Pollut. Rep.* **2022**, *8*, 273–287. [CrossRef]