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Changes in Wood Plastic Composite Properties After Natural Weathering and Potential Microplastic Formation

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Abstract: Wood plastic composites (WPCs) have recently gained attention as alternatives to traditional wood materials for outdoor use, thanks to their enhanced moisture resistance and durability, which extends their service life. Discolouration as well as surface erosion has been observed during weathering for both WPCs with untreated and heat-treated wood. However, aspects such as changes in surface hydrophobicity, chemistry, and erosion in terms of microplastic formation have received less attention; this research aimed to evaluate these factors during natural weathering. Four types of WPC samples, consisting of 50% wood particles (untreated and heat-treated) and 50% polypropylene, were naturally weathered in Latvia for two years. The samples measured 240 mm × 240 mm × 5 mm. Results showed rapid colour changes, microcracks, and exposed wood particles, suggesting microplastic formation. ATR-FTIR analysis showed increased absorption at 1715 cm^{-1} (carbonyl groups) and at 3410 cm^{-1} and 3460 cm^{-1}, typical of wood, indicating chemical changes on the surface. These changes influenced surface hydrophobicity, roughness, and water penetration. In a relatively short exposure time, WPCs without proper additives undergo significant changes in their aesthetic and physical properties, leading to surface erosion and potential microplastic formation. This could challenge the perception of WPCs as environmentally friendly materials.

Keywords: wood plastic composites; microplastics; weathering; natural weathering; polymer; environment

Citation: Vasiljevs, L.O.; Zabarovska, R.; Gulevska, E.; Cirule, D.; Sansonetti, E.; Andersone, I.; Andersons, B.; Kajaks, J.; Kuka, E. Changes in Wood Plastic Composite Properties After Natural Weathering and Potential Microplastic Formation. *Forests* **2024**, *15*, 2102. https://doi.org/10.3390/f15122102

Academic Editor: Antonios Papadopoulos

Received: 16 October 2024
Revised: 12 November 2024
Accepted: 22 November 2024
Published: 27 November 2024

1. Introduction

Wood plastic composite (WPC) production and demand are increasing all over the world due to their low maintenance requirements and high durability compared to wood [1]. Aesthetic appeal is another benefit considering that different colours and textures, including wood texture, can be made. In addition, they have a positive environmental impact due to the use of wood residues and recycled plastics in their production. By using WPCs, energy efficiency can be improved, waste can be effectively utilised, and biogenic carbon can be stored in the wood shavings, preventing CO_2 release into the atmosphere [2,3]. WPCs are used in many industries, such as construction, landscaping, packaging, and consumer goods. As of 2022, the industry of WPCs has an estimated worth of around USD 7 billion and is expected to reach USD 20 billion by 2035 [4]. In addition to wood, the second main component of WPC is thermoplastic polymers (polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS)), which are produced from fossil fuels and accounted for 4% of the total GDP of Europe in 2022 [5]. With Europe's plan to reduce 90% of net greenhouse gas emissions by 2040 and concerns about plastic waste accumulation in the environment, it makes sense to recycle more and utilise recycled plastic for the production of WPC [3,6]. This has already started in developed and developing countries in the 1990s, and in the last couple of years, the extent has only increased due to plastic pollution and the circular economy [7,8]. Despite that, WPCs are still predominantly manufactured from virgin plastic material [9].

Although WPCs are a viable alternative to other materials, they also have a few drawbacks that may turn consumers away from them. The main drawbacks of WPCs are their discolouration and loss of mechanical properties due to photo-oxidation [10]. Photo-oxidation refers to the breakdown of materials when they are exposed to sunlight. Sunlight consists of ultraviolet, visible, and infrared radiation, with UV radiation being the primary cause of polymer degradation. UV radiation is divided into three categories: UVA (320–400 nm), UVB (280–320 nm), and UVC (100–280 nm) [11]. Further, 95% of solar radiation consists of UVA, 5% consists of UVB, and UVC is almost entirely absorbed by particles in the upper part of the stratosphere [12]. Despite making up only a fraction of the total solar radiation, UVB is the main type of radiation that triggers the photo-oxidative degradation of common polymers [13].

Previous studies that investigated the changes in WPCs after exposure to natural and artificial weathering have reported whitening/fading and crazing. Whitening is the typical way in which WPCs discolour, but with prolonged exposure, a reversing effect can be observed, i.e., the colour of the material starts to darken due to the growth of mildew and mould on the surface [10,14]. The rate of discolouration can depend on various factors, such as environmental conditions (intensity, amount of precipitation, temperature etc.), wood filler characteristics (species, prior modification, extractives, etc.), polymer type, and additives (pigments, UV stabilisers, UV absorbers, antioxidants, etc.) [15,16]. Discolouration is generally linked to the photodegradation of both the polymer and the wood [15]. The incorporation of UV absorbers hindered amine light stabilisers, and pigments to a certain extent can protect both of the components, thus reducing the colour change. However, their efficiency decreases in time due to erosion, leaching, migration, photooxidation, etc. [17,18].

Crazing, which is the process of small crack-shaped region formation, results from chain scissions of the tie chain molecules in the amorphous phase of the polymer-releasing molecules, which then can crystallise onto pre-existing crystals (chemi-crystallisation) [19]. Chain scission is initiated by photo-oxidation [10,20,21]. In terms of chemical composition, the signal intensity for carbonyl groups (1680–1800 cm^{-1}) had increased after weathering but decreased for bands associated with methyl groups and tertiary carbons. Additionally, the bands attributed to cellulose at 3400 cm^{-1}, corresponding to free -OH groups, and between 1034–1108 cm^{-1}, associated with C-O and C-C stretching, exhibited an increase. Another study reported that after 240 h of artificial weathering, WPC samples (40 wt% wood flour and 60 wt% polypropylene) showed a decrease in contact angle values, which then increased after 960 h. The initial decrease was due to the photodegradation of polymer matrix and cellulose protrusion, while the subsequent increase was attributed to greater surface roughness and a polypropylene-rich surface [22]. All these results suggest that prolonged exposure to UV irradiation can significantly change the surface properties of WPCs, leading to erosion and the potential formation of microplastics (MPs). Given the complexity of the material, where both polymer and wood are subject to degradation and eventual erosion, focused research is essential in this area. This should involve a comprehensive approach using visual, chemical, and physical testing to gain a deeper understanding of surface changes, interpret the observed responses, and assess the potential release of MPs during the degradation process.

MPs have gained significant attention in society due to their harmful effects on the environment and human health. According to the ISO 21960:2020 standard, solid plastic particles with a size between 5 mm and 1 mm are classified separately as large MPs and particles with a size between 1 mm and 1 µm as MPs [23]. Particles smaller than 1 µm are classified as nanoplastics. Such small particles can pose a threat to humans and other organisms since they are less detectable and can enter the body more easily. In addition, plastics usually contain different additives, i.e., pigments, UV stabilisers, colourants, plasticisers, fire retardants, lubricants, coupling agents, etc., which can be harmful to living organisms as the initial application of plastic products does not consider the possibility of such an exposure. It has been estimated that mostly 69–81% of MPs are created via fragmentation from large plastics due to physical, mechanical, biological, or chemical degradation [24]. The main accelerators for the formation of MPs are abrasion, UV radiation, and heat, as well as different biological factors. These are the conditions to which construction materials, including WPCs, are exposed regularly during the service. The use of additives can significantly mitigate the degradation of plastics; however, it cannot completely prevent it due to leaching, erosion, photolysis, and photooxidation of the additives [25]. This eventually leads to the same unprotected products, and if not recycled or disposed of before this point, the degradation starts with similar outcomes as for the unprotected systems. As the MP question has hardly been studied for WPCs, it is important to analyse the changes in the surface leading to MP separation by applying the worst-case scenario, which is without additives. It also provides information on the consequences for products intended for indoor use, such as indoor furniture and reusable cutlery products, when they are exposed to the outdoor environment through littering or improper use. The market for these products is growing [26]. A contribution to the growth can be attributed to the European Union's "Plastics Directive" (2019/904), which aims to reduce the environmental impact of single-use plastics, prompting a shift in the production of disposable items, e.g., cutlery. To comply, manufacturers are turning to multi-use products and alternative materials, one of which is WPC [27]. Unfortunately, these materials still face scrutiny due to their non-biodegradable plastic content. In addition, as they are marketed as reusable, they are expected to last for some time, but there are no obligations for the products to be protected against UV radiation by the addition of suitable additives, and when added, the amounts are limited due to the regulations concerning food-contact materials. This limited UV protection can reduce durability in the outdoor environment. One more concern is that consumers may mistakenly assume these products as biodegradable due to their wood content, potentially leading to careless disposal.

Based on the current knowledge, it is believed that WPCs are environmentally friendly materials; however, they have not been evaluated from the perspective of MPs. Considering that WPCs partly consist of plastic (typically 30–70%) and are mainly used outdoors, it is very likely that they can be a source of MPs. These aspects have received little attention in the literature. The objective of the study is to evaluate the risk of MP formation during natural weathering by analysing the changes on the WPC surface.

2. Materials and Methods

2.1. Materials and Sample Preparation

Four types of WPCs with different compositions (Table 1) were made, each consisting of 50 wt% pine wood particles and 50 wt% polypropylene, with no additives added.

Table 1. Wood plastic composite composition (wt%).

Abbreviations	**Pine Wood Particles**			**Polypropylene**	
	Untreated	**Heat-Treated (Mild)**	**Heat-Treated (Moderate)**	**Virgin**	**Recycled**
UT-PP	50	-	-	50	-
HT1-PP	-	50	-	50	-
HT2-PP	-	-	50	50	-
HT2-rPP	-	-	50	-	50

Pine (*Pinus sylvestris* L.) wood particles were obtained from untreated and heat-treated (HT) wood. HT wood particles have been included in the study due to their superior durability against fungi and enhanced water resistance compared to untreated wood particles [28]. Accelerated weathering tests have shown reduced signs of surface erosion for WPCs with HT wood; however, data on natural weathering is not available. Further-more, WPCs containing wood particles modified at various heat-treatment conditions have different properties, and therefore not all WPCs made from HT wood will act equally during weathering. For evaluation of these differences, one untreated and two HT wood particles modified at mild and moderate conditions were included. The heat treatment was performed in a closed reactor system in a water vapour medium where the maximum pressure was 6 or 8 bar depending on the peak temperature. The peak temperature of the mild heat treatment was 160 °C with a holding time of 1 h at this temperature, and for the moderate treatment, it was 170 °C for 3 h [16]. Untreated, mild, and moderate heat-treated wood were separately milled through a 1.0 mm sieve to obtain wood particles.

For the production of WPC samples, commercial polypropylene (Mosten MA-712, "ORLEN Unipetrol RPA", Prague, Czech Republic) and recycled polypropylene were obtained and used from the local recycled plastic manufacturer "Nordic Plast" in Jelgava, Latvia. Recycled plastic was used only for the WPCs made of moderated HT pine as the most perspective composition in terms of durability. Prior to compounding, the wood particles were dried in an oven at 103 ± 2 °C for 24 h. The WPC material was then prepared using a LabTech two-roll mill with roll temperatures set to 180 °C and 185 °C and rotational speeds of 10 rpm and 20 rpm, respectively. After the compounding, the material was pelletised for further processing. Finally, the WPC samples were produced using compression moulding (t = 180 °C, p = 3.5 MPa) in hydraulic press Joos LAP 450 ("Gottfried Joos", Pfalzgrafenweiler, Germany), resulting in samples with dimensions of 240 mm × 240 mm × 5 mm.

2.2. Natural Weathering

WPC samples were exposed to natural weathering in Riga, Latvia, for 22 months starting in May 2022. The weathering experiment setup is presented in Figure 1. The samples were faced to the south with half embedded in the ground (unexposed part). All WPC samples were tested at three different weathering intervals: unweathered, after 11 months, and after 22 months. Both the south-facing and the north-facing sides of the part above ground were examined.

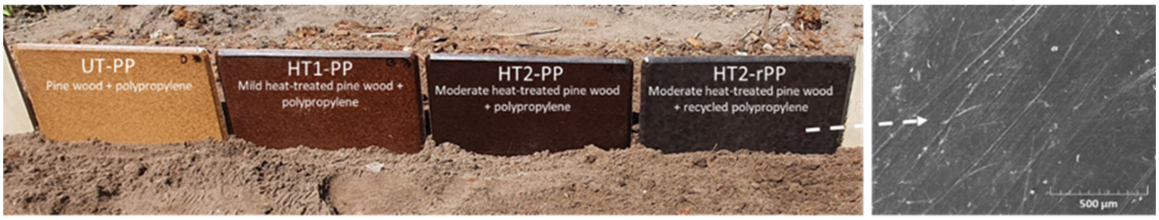

Figure 1. Experimental setup and scanning electron microscope image of unweathered surface.

The average values of weather parameters (temperature, precipitation, humidity and solar radiation) for each month during weathering were taken from a nearby meteorological station [29]. These data are summarised in Figure 2.

2.3. Colour Change

The colour before and after weathering was quantified by using a Konica Minolta CM-26dG spectrophotometer ("Konica Minolta", Tokyo, Japan). The measured parameters were L*, a*, and b* and the viewing angle was D65/10° with the specular component included. The CIELAB colour space system, being perceptual, facilitates the calculation of colour differences due to its uniform construction. The coordinates L*, a*, and b* are derived from reflectance curves and denote both lightness and chromaticity. An elevation in L* (ranging from 0 to 100) indicates a brightening of the material. The values of a* and b* represent chromaticity positions along axes spanning from −300 to +300, delineating the spectrum from green to red and from blue to yellow, respectively [30]. Both the south-facing and the north-facing sides were analysed, each side with six fixed locations where measurements were taken. Two surfaces for each type. The total colour change (ΔE_{ab}) was calculated according to Equation (1), where $\Delta L^* = L_2^* - L_1^*$, $\Delta a^* = a_2^* - a_1^*$, and $\Delta b^* = b_2^* - b_1^*$ with L_2^*, a_2^*, and b_2^* being the coordinates of weathered samples and L_1^*, a_1^*, and b_1^* being the coordinates of unweathered samples.

$$\Delta E_{ab} = \sqrt{\left(\Delta L^2 + \Delta a^2 + \Delta b^2\right)} \tag{1}$$

A more practical approach to recording changes in the colour of WPCs was executed by capturing images of the samples in a "white box" under the same artificial lighting conditions.

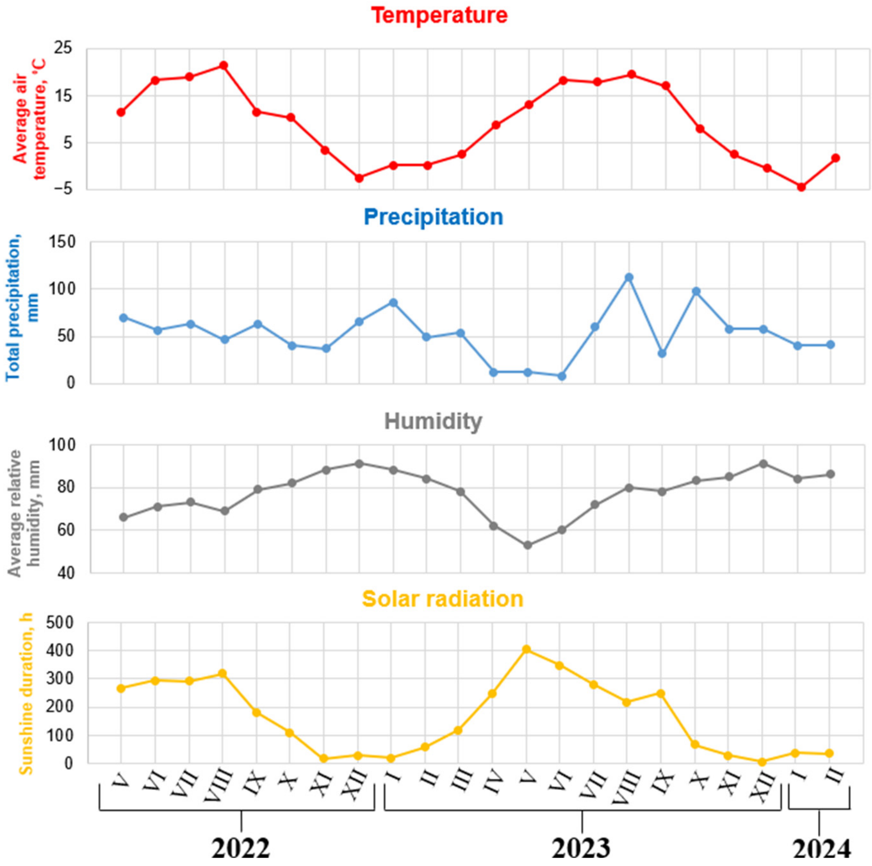

Figure 2. Average values of temperature, precipitation, humidity, and solar radiation each month from May 2022 until February 2024.

2.4. Surface Chemistry

Fourier transform infrared (FTIR) spectroscopy was conducted on a Nicolet iS50 FTIR Spectrometer ("ThermoFisher Scientific", Waltham, MA, USA). For each sample, 32 scans were performed in the absorbance range from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. Spectra were obtained using attenuated total reflectance (ATR) with a diamond crystal. For each combination of WPC type and weathering time, three replicates were analysed, with both the north-facing and the south-facing sides of each sample being examined. Due to the procedure of ATR-FTIR measurement and destruction of the surface during the process, the samples for the ATR-FTIR were obtained by cutting them out from the surface at different locations with a sharp razor after each weathering period. All spectra bands were normalised using the band at 2921 cm^{-1} that corresponds to CH_2 symmetric and asymmetric stretches in polypropylene.

2.5. Surface Hydrophobicity and Roughness

Surface hydrophobicity was evaluated by contact angle (CA) measurements, which were performed using a DataPhysics OCA 20 contact angle goniometer ("DataPhysics Instruments GmbH", Filderstadt, Germany). Drops of distilled water (10 µL) were dispensed onto the surface of the WPC at a deposition speed of 2 µL/min at six different locations, spaced 10–20 mm apart. The right and left angles of the drop on the surface were recorded, and the average of the two angles was calculated. CA was measured every second over a total duration of 30 s. The CA was determined by extrapolating the linear portion of the graph. The K value describes the penetration rate of the drop. It is calculated for CA values after reaching a constant wetting rate angle, as shown in Equation (2). For a method to calculate the constant wetting rate angle, see Nussbaum [31].

$$\text{K value} = \delta(\text{CA})/\delta\text{t} \tag{2}$$

Surface roughness was measured using a Mitutoyo SJ-210 ("Mitutoyo", Kawasaki, Japan) (standard type). The parameters set were: measuring speed of 0.5 mm/s, cut-off lengths λ_c of 0.8 mm and λ_s of 2.5 µm, auto range, and five sampling lengths. Measurements were taken at 10 different locations for each sample.

2.6. Surface Microstructure

A more detailed analysis of surface changes in WPC samples after weathering was performed using a scanning electron microscope (SEM) TESCAN VEGA TS 5136MN ("TESCAN", Brno, Czech Republic). The surfaces were sputter coated with gold and observed at 15 kV.

2.7. Statistical Method

To determine if there is a statistically significant difference between the two groups of values, an ANOVA (Analysis of Variance) test was conducted at a significance level of 0.05 in Excel. In this analysis, the *p*-value indicates whether the observed differences between groups are statistically significant, with a *p*-value less than or equal to 0.05 pointing to statistically significant differences between the analysed groups.

3. Results and Discussion

3.1. Discolouration

The results (Figure 3) show that after 11 months of exposure to natural weathering, the surface colour of all WPC samples became pale white due to photodegradation of the polymer, which is caused by surface crazing [16]. After 22 months, the colour of the surfaces facing south darkened, likely due to microbial growth and surface erosion, which led to the separation of MPs and exposed the darker wood particles beneath the weathered surface. Similar colour changes have been noted in previous studies, where the surface was initially whitened after a few months of exposure [32,33]. However, with further exposure, the colour stabilised and eventually began to darken [32]. These observations were detected both visually and through SEM investigations, which are discussed later in this paper. The colour changes after 11 months of exposure are more pronounced on the south-facing sides. However, the north-facing sides also exhibited statistically significant ($p < 0.05$) colour changes, despite receiving only reflected and less intense UV radiation.

The changes in colour parameters (L*, a*, b*) in the CIELAB colour space are presented in Figures 4 and 5. The results show that the south-facing side whitens after 11 months of exposure to natural weathering, regardless of the relatively light initial colour. In some studies, the whitening process is explained by the photodegradation of lignin, which produces water-soluble groups that leach into rainwater, exposing brighter-toned cellulose [30]. This explanation is most likely adapted from wood research and its greying mechanism. However, in our previous study, it was identified that the whitening of WPCs is actually caused by the microcracking of the polymer matrix, showing a strong correlation between the colour change (whitening) of WPC and the microcracking of the polymer matrix that reduces visible light transmittance [16]. After an additional 11 months of weathering, the WPC surface becomes darker, which indicates that the separation of MPs from the surface of the material has occurred, exposing darker wood particles. Microcracking and formation of MPs could be explained by the photodegradation mechanism that occurs via Norrish type I and results in the formation of free radicals that cause polymer chain scissions, reducing the molecular mass and strength [15]. Alternatively, degradation can occur through the Norrish II reaction, which leads to the formation of carbonyl groups and terminal vinyl groups while also causing chain scission. Thus, during accelerated photodegradation, both chain scission and crosslinking mechanisms are at play [34]. The process that can facilitate the formation of MPs is the swelling and shrinkage of wood particles; however, further research is needed in this direction. The change in L*, a*, and b* parameters of the north-facing side after 11 months of weathering (Figure 5) shows a much slower shift in colour parameter values. Another 11 months of exposure changed the surface colour to white, but not as white as the south-facing side after the first 11 months of weathering. The differences in the initial colour of the samples due to various wood and polypropylene types had no effect on the end colour, as for all samples it shifted from the initial colour to white.

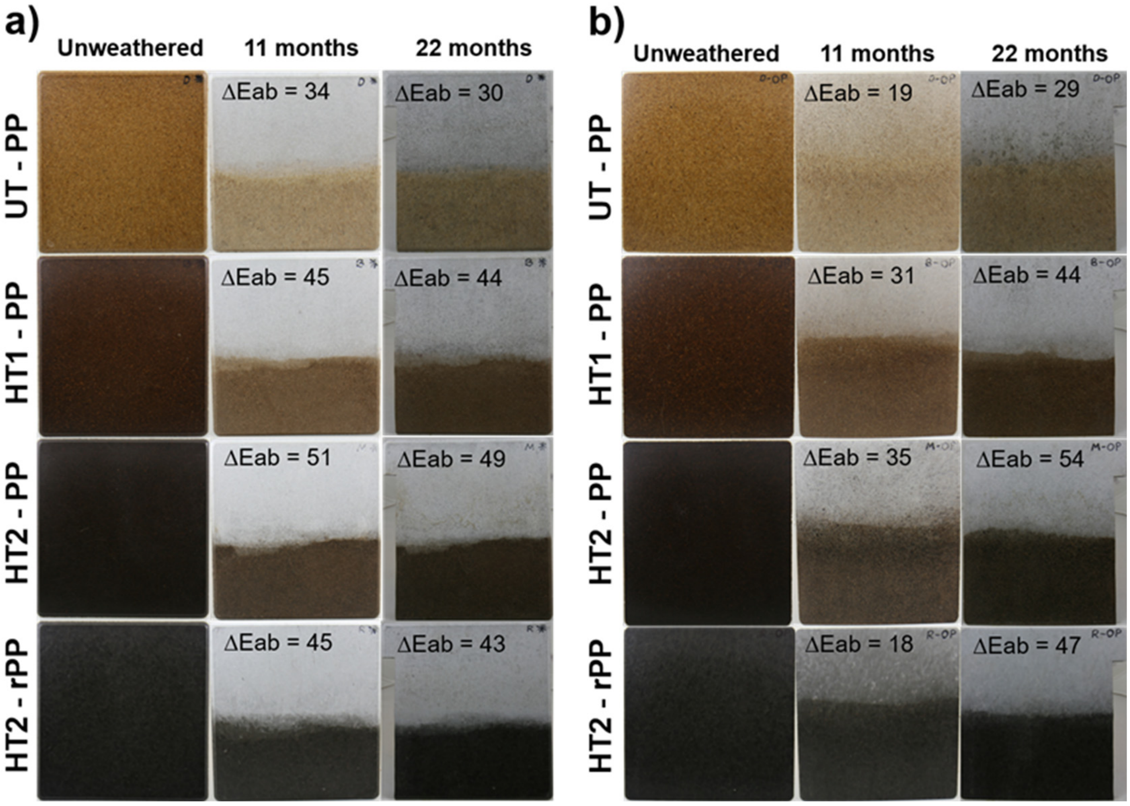

Figure 3. Changes in surface colour of WPC samples: (**a**) South-facing side; (**b**) North-facing side. The bottom half of the sample was embedded in the ground and remained unexposed throughout the test—for this part, the colour changes were not evaluated.

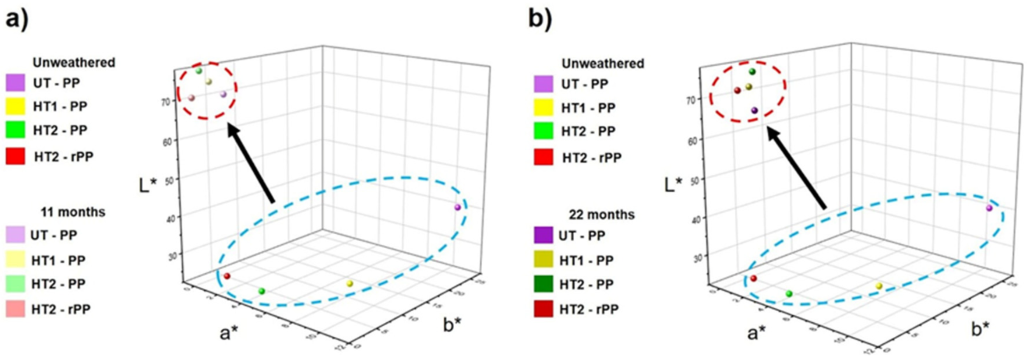

Figure 4. Changes in L*, a*, b* parameters for the surface of WPCs south-facing side: (**a**) Unweathered surface in comparison with 11 months of exposure to natural weathering; (**b**) Unweathered surface in comparison with 22 months of exposure to natural weathering.

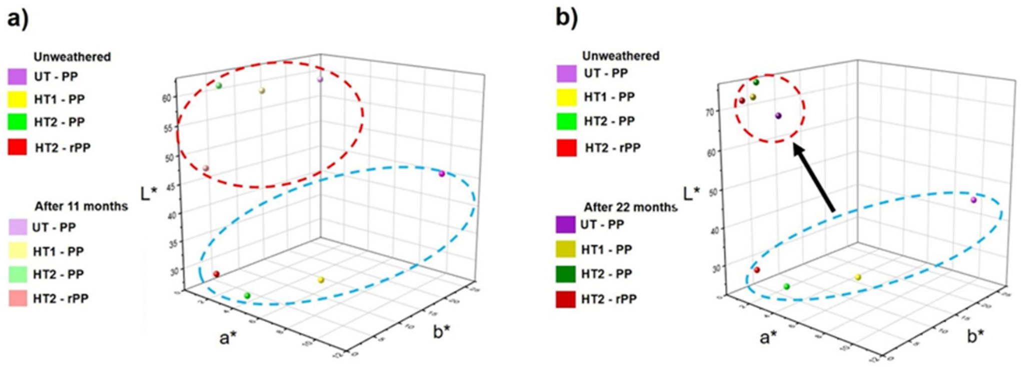

Figure 5. Changes in L*, a*, and b* parameters for the surface of WPCs north-facing side: (**a**) Unweathered surface in comparison with 11 months of exposure to natural weathering; (**b**) Unweathered surface in comparison with 22 months of exposure to natural weathering.

3.2. Chemical Changes

The given WPCs consist of wood particles and polypropylene, each with distinct FTIR bands. To understand the expected results, we initially captured the spectra of polypropylene and wood separately (unweathered and after 500 h of UV irradiation) (Figure 6). The bands of most interest were at 3410–3460 cm^{-1}, 1740–1720 cm^{-1}, 1500–1510 cm^{-1}, and 1031 cm^{-1}. The broadband from 3410 to 3460 cm^{-1} is associated with hydroxyl groups in phenolic and aliphatic structures, usually found in lignin and cellulose. The band at 1500–1510 cm^{-1} is attributed to the aromatic C-H vibrations of lignin. The band at 1740–1720 cm^{-1} corresponds to the C-O stretch in unconjugated ketones, carbonyls, and ester groups (frequently of carbohydrate origin), but the band at 1031 cm^{-1} can be equated to C-O stretching in cellulose and hemicellulose. This band is very pronounced in wood spectra, but there are no noticeable bands in this region in the polypropylene spectra [35]. The most prominent bands of polypropylene are in the 2800–3000 cm^{-1} range, corresponding to symmetrical and asymmetrical C-H stretches, and at 1377 cm^{-1}, which is associated with the CH_3 umbrella mode (Figure 6) [36].

WPC samples were analysed using FTIR spectroscopy before, after 11 months (Figure 7), and after 22 months (Figure 8) of weathering. The presence of a band at 1740–1720 cm^{-1} after weathering indicates an increase in carbonyl groups. Although both the polymer and wood are susceptible to photodegradation, the signal indicating the rise in carbonyl groups most likely originates primarily from the polymer. This is because the carbonyl products from wood are water-soluble and can be washed away by rain [37]. After 22 months of weathering, there is noticeable growth of the band at 1031 cm^{-1}, indicating that the polymer has detached from the WPC surface, allowing more wood characteristics to be seen in the spectra. Additionally, the increase in the band at 3410–3460 cm^{-1} further supports this observation, as this broadband is prominent in wood spectra. This change is not observed after 11 months of weathering, suggesting that the polymer had not yet detached from the surface at that point.

The only sample showing a slight increase in the band at 1031 cm^{-1} after 11 months of weathering is the UT-PP indicating higher erosion and more exposed wood particles. This may be attributed to the enhanced compatibility of TM wood with the polymer matrix and reduced swelling of the wood particles, which can otherwise lead to easier detachment of the polymer [16,38]. However, this characteristic is less noticeable after 22 months of weathering. Although TM wood delays the detachment of polymer particles, it does not completely prevent it.

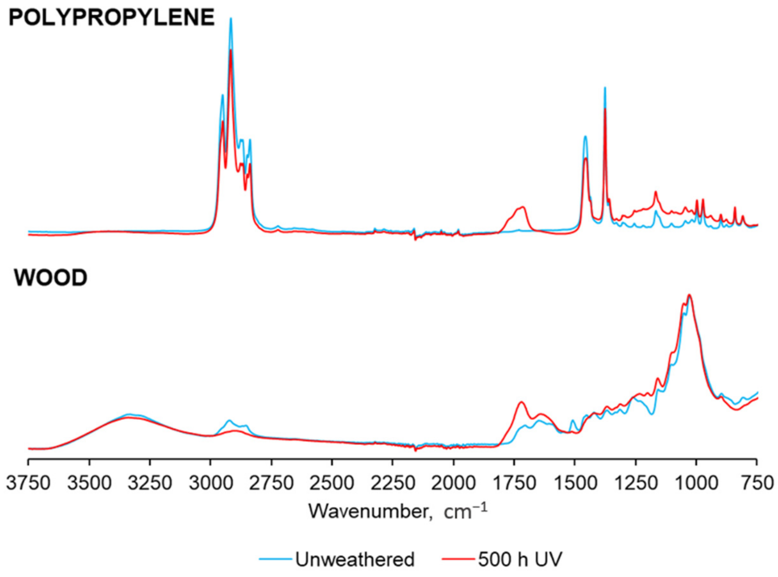

Figure 6. ATR-FTIR spectra of wood and polypropylene unweathered and after 500 h of UV irradiation.

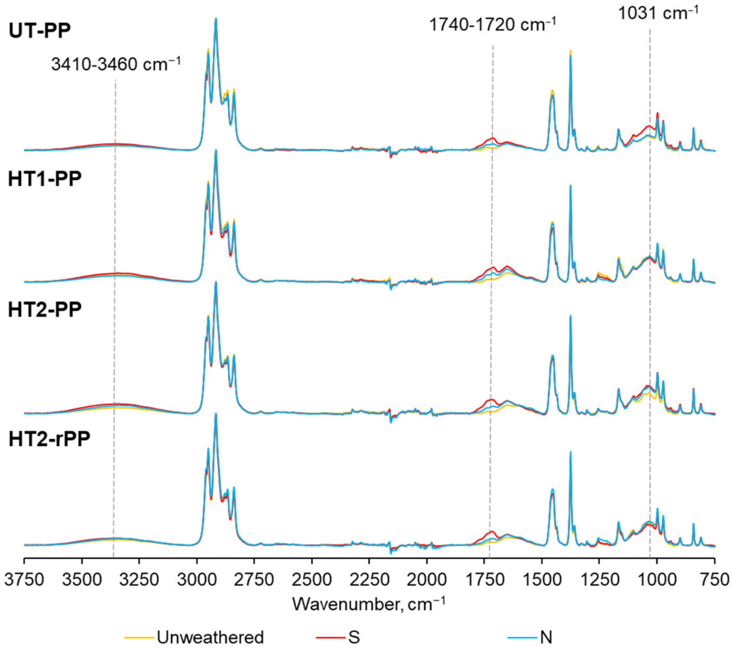

Figure 7. ATR-FTIR spectra of WPC surface (unweathered and after 11 months of weathering the north-facing (N) and the south-facing side (S)).

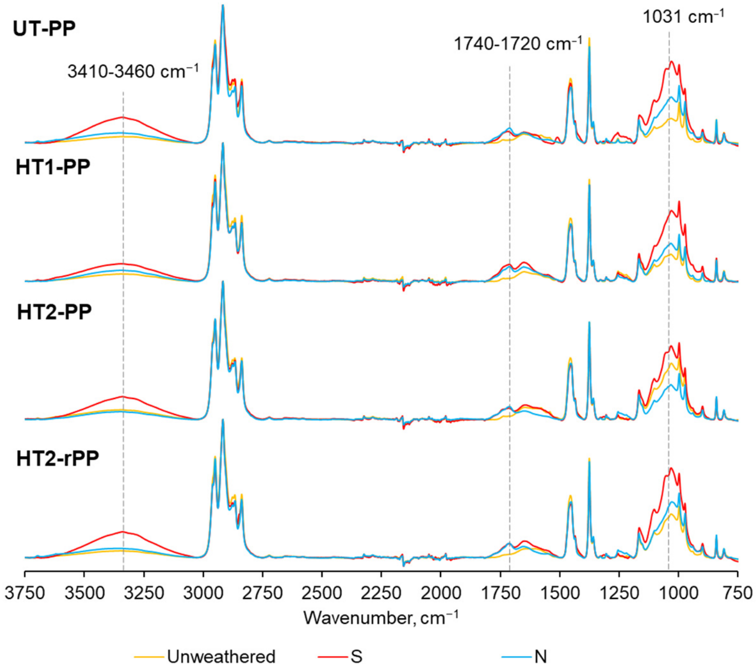

Figure 8. ATR-FTIR spectra of WPC surface (unweathered and after 22 months of weathering the north-facing (N) and the south-facing side (S)).

La Mantia et al. (2008) observed that as irradiation time increased, the spectra of polypropylene/wood flour samples showed a rise in the carbonyl band [39]. Similarly, Peng et al. (2014) identified an increase in carbonyl groups and additionally noted a rise in the bands corresponding to free -OH in cellulose (3400 cm^{-1}) and other characteristic cellulose bands (1034–1108 cm^{-1}, 1315 cm^{-1}, 1425 cm^{-1}) [22]. They suggested that this could indicate a greater presence of wood flour or cellulose fibre on the surface of the samples after weathering. Our results are consistent with previous findings.

3.3. Changes in Surface Properties

CA measurements reveal surface wettability, which is influenced by factors such as surface energy, roughness, chemistry, and sample preparation. While the polymer component of WPC exhibits hydrophobic characteristics, the wood component is hydrophilic. After 22 months of natural weathering, there was no statistically significant change ($p < 0.05$) in the CA of WPCs. Although ATR-FTIR results indicate an increase in functional groups that are specific to wood—which would typically suggest a lower CA—the CA remains unchanged, most likely due to an increase in surface roughness (Ra) (Figure 9) [22,38]. Peng et al. (2014) observed that the surface wettability of polypropylene composites reinforced with wood flour, lignin, and cellulose undergo continuous changes in CA at different stages of artificial weathering. They found that the protrusion of cellulose combined with surface erosion during weathering tended to result in decreased CA values. In contrast, the presence of lignin and the formation of a new layer of PP led to an increase in the CA [22]. However, surface erosion does not always result in decreased CA values. Wang et al. (2020) observed that as the surface roughness increased, the CA initially kept rising. It continued to increase until it reached its highest value. After hitting this maximum point, the CA angle began to decrease as the surface roughness continued to increase within the studied range [40].

To better understand the surface changes, the K value, which describes the water penetration rate, was calculated. The observed increase in the K value (Figure 10) after 22 months of natural weathering suggests surface erosion and microcracking of the WPCs. This finding correlates with an increase in surface roughness (Ra) (Figure 9) [41]. Microcracks in WPCs facilitate cyclic water absorption and desorption, leading to the material's shrinkage and swelling. A higher K value indicates increased intensity of these processes, which promote crack formation and propagation and infiltration of microorganisms, ultimately causing significant material degradation and a reduction in mechanical properties [38,40]. According to Defoirdt et al. (2010), moisture sorption can pose significant problems, leading to distortions, swelling, and buckling [42]. Additionally, the absorption of water can promote fungal growth, which was observed for the weathered WPCs, especially UT-PP [42,43].

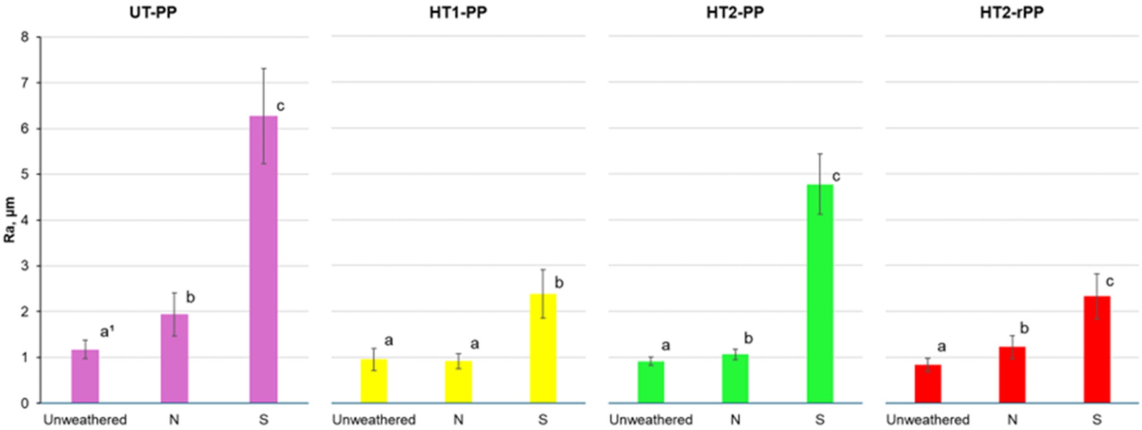

Figure 9. Ra value for the surfaces of WPCs (unweathered and after 22 months of weathering north-facing (N) and the south-facing side (S)). [1] Groups with the same letters in the column indicate that there is no statistical difference ($p < 0.05$) between exposure conditions within the sample series.

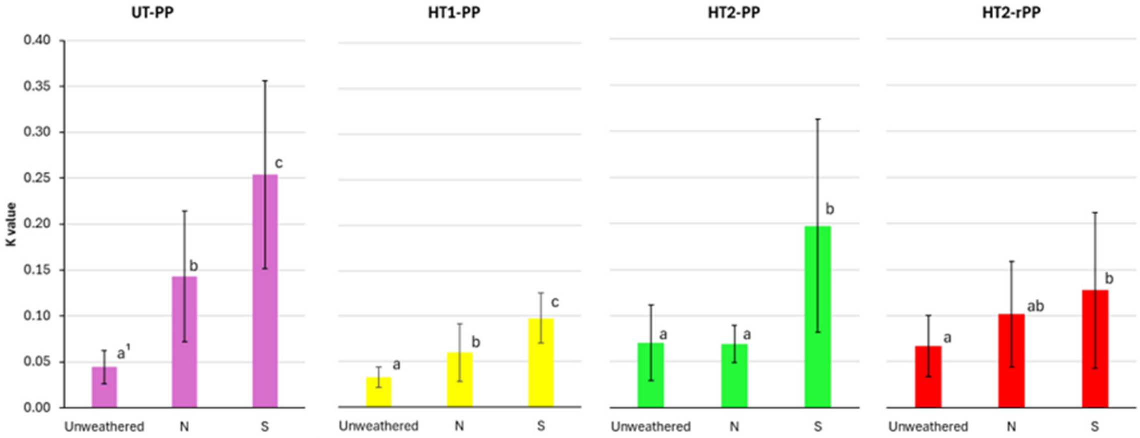

Figure 10. K value for the surfaces of WPCs (unweathered and after 22 months of weathering the north-facing (N) and the south-facing side (S)). [1] Groups with the same letters in the column indicate that there is no statistical difference ($p < 0.05$) between exposure conditions within the sample series.

The change in K value is less pronounced for WPCs containing heat-treated wood particles, resulting in reduced water sorption compared to WPCs with untreated wood particles. The degradation occurs more slowly, which can be attributed to improved compatibility with the polymer matrix and reduced swelling and shrinkage of the wood particles. Kuka et al. (2016) found that heat-treated wood fibres in WPCs absorb less water due to a decrease in hemicellulose content. This decrease in hemicellulose content makes the wood less hydrophilic and improves compatibility between the wood and the polymer matrix, rendering it less accessible to water [44].

3.4. Surface Morphology and Potential Microplastic Formation

The surface degradation of the weathered WPCs is shown in Figure 11. The SEM images correlate well with the results from the colour analysis, where it was concluded that the lightness of the WPCs decreased between the 11th and 22nd month due to the separation of MPs from the surface of the material, exposing darker-toned wood particles. Gunjal et al. reported similar results, indicating that the microcracks in the polypropylene (PP) composite became more pronounced after prolonged weathering [32]. The SEM images show that after 22 months, the crazing of the polymer matrix is more pronounced than after 11 months and more wood particles are visible due to the surface erosion separating the degraded plastic from the bulk of the material. These findings indicate that the eroded materials are definitely MPs, with a size of <500 µm for untreated pine wood and <300 µm for heat-treated pine wood. MPs of smaller size are considered more harmful due to increased absorbency [45], suggesting that WPCs with heat-treated wood could eventually produce more harmful MPs. However, considering the extent of the observed surface erosion, the number of formed MPs is much larger for WPCs with untreated wood. The differences in the size of the eroded MPs between WPCs made with untreated and heat-treated wood could be due to the differences in the light transmittance of the materials, resulting in different microcracking patterns [16]. The separation of MPs is triggered by the water penetration through these microcracks in the material and subsequent swelling of wood [46]. The FTIR spectra show an increase in absorbance for functional groups commonly associated with wood, which also correlates well with the SEM images. The microcracking and unveiling of the wood particles align well with the increase in K value and surface roughness (Ra). The formation of MPs is most likely driven by microcracks that allow water infiltration into the material, leading to swelling of the wood particles. This process induces internal stress and weakens the adhesion between the wood particles and the degraded plastic, ultimately resulting in erosion. Previous results have shown that water penetration can cause irreversible loss in adhesion by forming gaps between the wood particles and plastic [28].

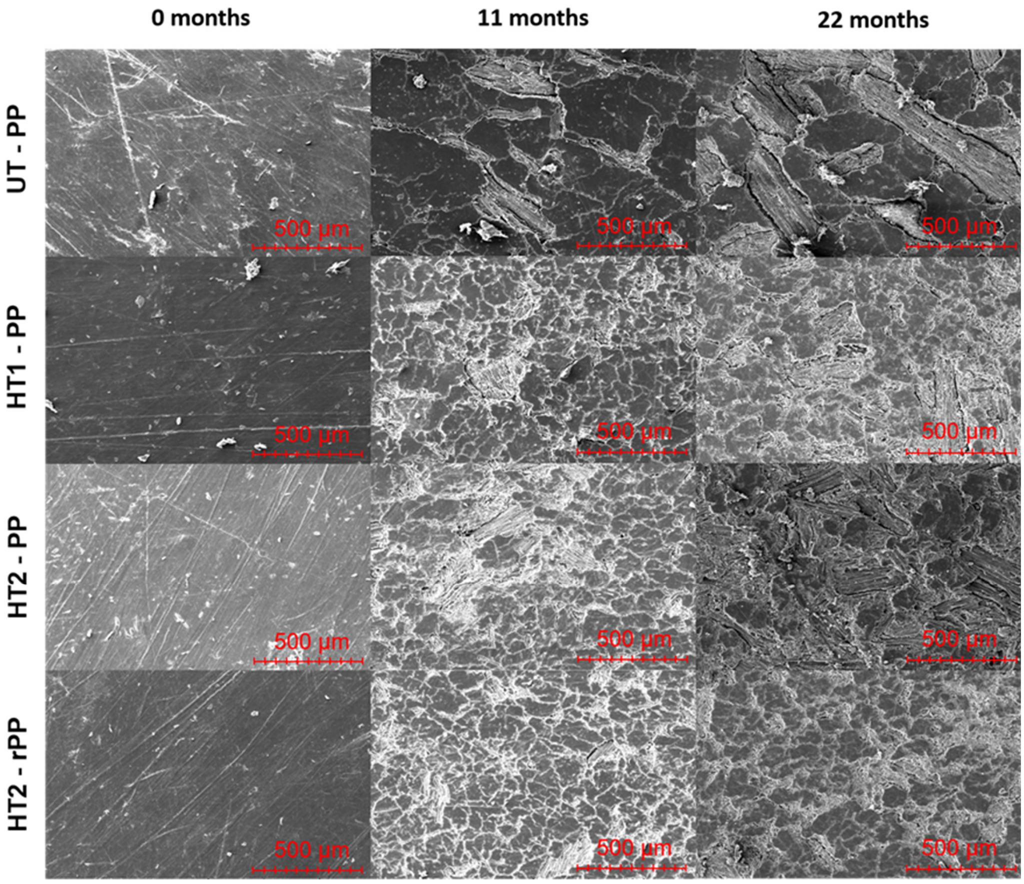

Figure 11. SEM images depicting the effects that natural weathering has on the surface of WPC.

4. Conclusions

The study showed that the approach used to investigate the formation of MPs from WPCs due to weathering can provide valuable insights into the visual, physical, and chemical changes associated with the process. The surface of all tested WPCs significantly degrades during weathering. WPCs containing heat-treated wood exhibit a slower degradation rate compared to those with untreated wood. However, they have a higher potential for releasing smaller MPs, which may pose greater environmental risks. Overall, the results suggest that WPCs can be a source of MPs; therefore, it is important to mitigate this risk by the use of suitable additives that can protect the polymer matrix from photodegradation. In addition, due to the degradation of additives over time, the risks of MP formation at later stages of the life cycle should be considered and studied also in the future to understand the risks if responsible collection and recycling or disposal of the material is not carried out. The key conclusions from the research are as follows:

- After weathering, the surfaces of WPCs exhibit significant degradation resulting in oxidation, embrittlement, and erosion causing discolouration, increased surface roughness, and water penetration, leading to further degradation possibly influencing mechanical and other material properties.
- The exposed wood particles on the surface of the WPCs, identified by SEM imaging and FTIR spectra, indicate that MPs have separated due to natural weathering, suggesting that these materials can be a source of MPs. Based on the observed microcracks, the size of the eroded MPs is less than 500 µm.

Although the surface of weathered materials was analysed, further research on MP formation from WPCs is essential. Techniques and methods that would allow analysing the eroded MPs are needed that would help to determine their quantity and type, providing valuable information for risk assessment and characterisation of MPs.

Author Contributions: The authors confirm contribution to the paper as follows: study conception and design: E.K. and D.C.; data collection: L.O.V., R.Z., E.G. and E.S.; analysis and interpretation of results: L.O.V., R.Z. and E.G.; draft manuscript preparation: L.O.V., R.Z. and E.G.; review and editing: D.C., I.A., B.A. and E.K.; resources and project administration: J.K., I.A. and B.A. All authors reviewed the results and approved the final version of the manuscript. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Latvian Council of Science project MicroWPC, project No. lzp-2022/1-0639 and by the Latvian State Institute of Wood Chemistry Bioeconomy Grant WoodREx project No. 10-24.

Data Availability Statement: Data are available on request from the authors.

Conflicts of Interest: The authors declare no conflicts of interest.

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