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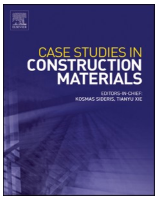

Risk of rainfall caused leaching from bio-composite material based building façades into the aquatic environment

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ABSTRACT

The increasing focus on sustainability and circularity is driving the global production of environmentally friendly products. The Netherlands started producing new bio-composite materials which are created by reclaiming resources from various sectors of the water industry. These materials can be used for a variety of applications including façade elements in buildings. However, their potential environmental impact, particularly with regard to leaching of potentially harmful substances into surface water, necessitates further evaluation. To address this issue, a systematic environmental risk assessment methodology combined with novel experimental data is presented here. To collect this data, façade panels made of two different bio-composite materials were first subjected to a series of laboratory tests, including analysis in both new and weathered forms, the latter subject to a cyclic UV radiation and high humidity, in order to simulate the effects of aging. Leaching tests were then conducted to determine the potential release of specific chemical substances such as heavy metals and resin compounds, under two different rainfall conditions (every day and more extreme). The data generated this way was used to perform the risk assessment using the existing European ERA framework. The results obtained reveal different leaching behaviour of the new and weathered samples, as well as between the two analysed bio-composite materials, depending on the rain intensity. To overcome the uncertainties caused by the limited input data, a sensitivity analysis was carried out whereby leaching concentrations and rainfall intensities were varied and their influence on the environmental risk was assessed. The results obtained demonstrated that, despite some variability, both materials appear safe to use, i.e., with estimated risks below the established safety threshold. While these findings provide a preliminary indication, they are based on laboratory conditions and assumptions hence further field studies are recommended to obtain more definitive conclusions.

1. Introduction

The climate crisis is becoming a priority for most of the industries globally, particularly in Europe. There is a growing inclination towards the utilization of environmental-conscious solutions, renewable resources, and materials with a reduced environmental impact, which is leading to an enhanced market value and a reduction in greenhouse gases emissions [1].

The use of sustainable and green materials is becoming increasingly prevalent in a range of applications, including water industry

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[2,3], packaging, the automotive industry, agriculture, and medicine [4]. A novel bio-composite material, made from water resources, is now being manufactured in the Netherlands. However, the utilization of these innovative materials, despite their derivation from natural resources, may give rise to concerns pertaining to environmental risk due to the potential chemical contaminants introduced by the raw materials employed in their production. The prolonged exposure of materials to environmental factors, including sunlight, rain, snow, and wind, may result in a deterioration of their quality. This could give rise to concerns regarding the potential release of pollutants or an increased demand for environmentally intensive replacement.

The subject of this study is the new bio-composite materials, which are produced from resources recovered from the water cycle as described by Nativio, Kapelan [5]. It is likely that the aforementioned water sources are likely to be contaminated with heavy metals, including Ba, B, Mn, and Zn [6], which may pose a risk to both human health and the environment. Moreover, bio-composite materials, like other composite materials, utilize resins such as polyester-based or furan resins, which contain hazardous substances, including styrene and furfuryl alcohol. The novel bio-composites are primarily employed in aquatic settings, serving as water level indicators and canal bank protection elements. Nevertheless, they are also being employed in the construction industry as façade elements.

The primary concern regarding traditional façades, as evidenced by the scientific literature and industry regulations [7], is the leaching of biocides from the façade during precipitation events. Biocides are defined as substances designed to inactivate or inhibit the growth of algae, fungi and other microorganisms via chemical or biological reactions as outlined in the European Directive 98/8/CE [7,8]. It is frequently observed that plasters, mortars, and coatings utilized for external thermal insulation and finishing protection frequently contain water-soluble biocides that migrate within the coating and subsequently leach into the surrounding environment over time. In order to evaluate the leaching of biocides from façade building elements, both outdoor and indoor tests were conducted. Schiopua, Tiruta-Barnab [9], with the aim to meet the standards in the European Directive concerning the placing of biocidal products on the market [7], developed a chemical transport model of the leached chemicals during rain events. Laboratory tests were conducted using a mathematical model and software to simulate the precipitation scenarios. The chemical transport model, developed at laboratory scale, was implemented by incorporating external parameters pertaining to natural exposure conditions. Subsequently, the laboratory scale indoor tests were then compared to the outdoor tests, which consisted of exposing samples to the outdoor environmental conditions. The simulation conducted in this study demonstrated the feasibility of modelling leaching behavior in a satisfactory manner [9]. In a study conducted by T.P. Wangler, S. Zuleeg [10], it was observed that the release of biocide from building façade elements was diffusion controlled under controlled wetting and temperature conditions. Furthermore, an increase in ambient temperature was observed to directly corresponded with an increase in emissions rate. The orientation of the façade was identified as significant factor influencing leaching, as demonstrated by Vega-Garcia, Schwerd [11]. In their study, the researchers investigated the correlation between the leaching of biocides and the wind driven rain (DWR), which was identified as the primary parameter influencing the leaching load.

In addition to the potential leaching of biocides, the leaching of other potentially dangerous substances, such as heavy metals, is also a cause of concern. Islam, Jo [12] conducted column leaching tests to investigate the leaching and mobility of copper (Cu) and lead (Pb) due to simulated road runoff. In accordance with this, runoff may be responsible for the leaching of heavy metals and could also result in groundwater contamination by transferring heavy metals leached in the soil to the groundwater. The aggregation of toxic elements in the ecosystem is a significant consequence of heavy metals accumulation, which can occur through various pathways, including plant root uptake. The accumulation of heavy metals can have adverse effects on the food chain, which may result in acute and chronic health issues in humans [13]. Weiler and Vollpracht. [14] investigated the potential leaching of heavy metals and hazardous substances from concrete carbon composite façade elements. The evaluation was conducted through the performance of outdoor and indoor tests, with the objective of assessing the validity of laboratory experiments and determining their applicability in an outdoor setting. No correlation was observed between laboratory leaching data and outdoor emissions. This lack of correlation was mainly dependent on the substance leached and the method used. Additionally, the wetting cycle of the laboratory tests needs to be optimized to better simulate outdoor conditions and represent outdoor leaching behavior more accurately. However, laboratory leaching tests could be used to vary single factors and better understand outdoor leaching behavior.

As can be seen from above, the optimal conditions for conducting leaching tests on building facades are in a field or outdoor setting. Nevertheless, this would be challenging and costly to accomplish, for evident reasons. An alternative approach would be to conduct laboratory or indoor tests where rainfall events can be simulated under more controlled conditions. Nevertheless, it is not straightforward to achieve this, as accurately reproducing the intricacies of outdoor meteorological conditions within a laboratory setting presents a considerable challenge in itself. For instance, raindrop size is a pivotal factor of the crucial parameter since it has an effect on the drop velocity and consequently on the kinetic energy which affects the detachment process [15]. To simulate precipitation events in an indoor setting and evaluate droplet size, rainfall events were simulated both using pressurized nozzles (which employ a gravity-based flow rate) and no-pressurized nozzles (which utilize an upstream pressure-based flow rate) [16,17]. Weiler and Vollpracht. [14] refer to the common mean diameter of raindrops in Western Europe, specifically in Germany, which is approximately 2.4 mm, with the objective of replicating precipitation intensities within the range of 1–5 mm/h. It was also noted that in order to accurately simulate realistic rain intensities, the droplet size employed in the experiments (in terms of droplet diameter) was smaller than the actual size that the drops would have in reality, due to the limitations of the kinetic energy representation [18]. The complexity of indoor experiments, encompassing variables such as water head, velocity, and droplet shape, renders an accurate representation challenging to achieve [18]. It is evident that laboratory-based leaching tests with simulated rainfall are challenging to conduct and that these cannot fully substitute for outdoor leaching tests. Nevertheless, indoor leaching tests can serve as a valuable preliminary assessment of environmental risk potential, providing initial insights before the experiments are conducted outdoor.

The new bio-composite materials used in this work have previously been analyzed in terms of environmental risk, but in a different application namely, as canal bank protection elements [19]. However, it should be noted that an environmental risk assessment has yet to be conducted for the façade elements under analysis, which present their own particular setoff characteristics and challenges. To address this knowledge gap, this study aims to develop and present an approach for risk assessment based on the leaching of materials from façade elements (made of new bio-composite materials) into the aquatic environment due to rainfall events. The environmental risk assessment is conducted in accordance with the established methodology [20] with newly collected input data obtained through the simulation of rainfall events under laboratory conditions and subsequent conduction conducting of corresponding leaching tests. Although the leached hazardous substances from façade elements have the potential to pollute the air, surface water, soil, and groundwater, this studyfocuses exclusively on the risk of surface water pollution.

The paper is structured as follows. Section 2 presents the methodology used to perform the leaching analysis, including the related experimental design. Section 3 presents the obtained results with relevant discussions. Conclusions are presented in Section 4.

2. Materials and methods

2.1. Overview

In this study the framework developed and tested in the previous study by Nativio, O. [19] was employed to evaluate the leaching of components from bio-composite materials utilized as façade elements. The framework is structured into three principal blocks: (i) the existing Environmental Risk Assessment (ERA) methodology [20] and the corresponding Dutch guidelines published by the Dutch National Institute for Public Health and the Environment (RIVM) [21]; (ii) the European Chemical Agency [22] database; (iii) laboratory-based sample preparation and leaching tests. Given the novelty of the bio-composite materials, on-site leaching data were not available for analysis. Consequently, laboratory leaching tests were employed to simulate the actual scenario and to predict the potential leaching over time. The application of bio-composites as façade building elements requires an evaluation of the potential for leaching following adverse weather conditions, such as rainfall events. In accordance with this approach, two precipitation events, each lasting one hour, were selected from the Koninklijk Nederlands Meteorologisch Instituut (KNMI) database [23] for evaluation of leaching behaviour in relation to rainfall intensity.

2.2. Bio-composite materials used as façade building elements

The study assesses the environmental risks associated with the potential leaching of heavy metals and resin compounds from façade building elements made from new bio-composite materials. The use of bio-composites as façade building elements is exclusively for aesthetic purposes.

Four distinct bio-composite alternatives have been produced in the Netherlands, and for the purposes of this study, two of them have been selected for analysis. The characteristics of the selected materials are presented in Table 1. The precise compositions and percentages of each raw material are not presented in this study due to the confidential nature of the data. However, all pertinent parameters were rigorously considered in the analysis and that the materials were tested in accordance with established protocols to guarantee the precision of the results. The materials are composed of natural fibres (reeds and grass clippings from the canal maintenance, wastewater cellulose) and fillers (calcite from the drinking water softening process and bio-filler from agricultural waste) with the addition of additives. The materials are bonded together using a resin (polyester-based resin, or furan resin), as detailed in Table 1.

The novel bio-composite materials are manufactured through the following process, as outlined by Nativio, O. [6]:

- The production of the material begins with the preparation of the dough. The resin (in a fluid state) is combined with the filler (e.g., calcite) and mixed for a period of 20 minutes. Subsequently, the fibers are incorporated into the batch and mixed for a period of 2–4 minutes. The fibers are introduced at the final stage of the process and combined for a shorter duration in order to prevent their structural integrity from being compromised. The mixing process facilitates the cohesion of all the raw materials, thereby ensuring compatibility and adhesion. In case of M4, the bulk molding compound (BMC), is subject to a heating process during the mixing stage, with the objective of reducing the water content.
- The obtained dough is vacuum sealed in bags and stored in a refrigerator for approximately one week prior to being pressed.

Table 1
Composition of bio-composite materials used as façade building elements.

Materials	M3	M4
Natural fibers	Wastewater cellulose	Grass
Filler	DW Calcite[a]	Bio-filler from agricultural waste
Resin	Polyester resin with bio-based content[b]	Furan resin[c]
Additive	Additives[d]	Additives[d]

[a] DW Calcite concerns the calcite pellets collected as residual from drinking water softening, one of the drinking water treatment processes in treatment schemes.

[b] Bio-based polyester resin has a lower amount of Styrene (~35 %).

[c] Furan resin containing furfuryl alcohol.

[d] Additives: used as catalyst to stimulate the chemical reaction (polymerization) of the resin, improve impact resistance (impact agents), release agents to prevent sticking to the mould and allow releasing the part (release agents). Zinc stearate as release agent for M1 and olive pamoate for M2.

- Finally, a molding process is applied with the objective of imparting the desired shape to the product (e.g., to create panels). The materials are placed beneath the press (for bulk molding process). The dough is weighed and fed into the mold, then the material is pressed for the required curing time before the part is removed from the mold.

The resulting product may be subjected to additional processing, should this be necessary for the specific application in accordance with the requisite specifications.

Two distinct samples of bio-composite types of façade elements were subjected to analysis for each material. The first type was the new sample. These samples were used to evaluate the leaching behaviour of new façade panels. The samples were produced approximately three months prior to testing and were stored under dry conditions and without sun exposure and are defined as ’new’ in this manuscript. The second type analysed were samples that after production underwent cyclic exposure to simulated solar irradiation and extreme humidity (100 % relativity humidity, RH), after production in order to accelerate the weathering and ageing effects. The aforementioned samples, which had undergone treatment, were designated as "weathered" in the course of this study.

The utilization of additives, particularly the release agent, has been observed to increase the prevalence of chemical contamination in terms of zinc (Zn) for M3 (where zinc stearate was employed as the release agent). Moreover, the study conducted by Quero, Ballesteros [24]indicates that the olive pamoate, employed as the release agent for M4, may be susceptible to contamination with chemicals elements such as calcium (Ca), potassium (K), magnesium (Mg), sodium (Na), manganese (Mn), and zinc (Zn).

2.3. *Weathering treatment of samples*

A total of four samples were analysed, with two samples (one new and one weathered) for M3, and two samples (one new and one weathered) for M4. The preparation of the weathered samples entailed the placement of new materials in an accelerated weathering tester, QUV/se by Q-LAB-UVA for a period of 1372 hours (i.e., just over 57 days). During this period, the samples were subjected to cycles of UVA irradiation (0.89 W/m^{-2} at 340 nm for a period of 8 hours at 60 °C) and humidity (100 % RH for a period of 4 hours at 50 °C), following ASTM-G1545 Standard [25,26]. In accordance with the recommendations set forth in the weathering machine guidebook [27] and utilizing the annual solar irradiation data for the Rotterdam area obtained from the Netherlands Meteorological Service database [23], it was possible to ascertain that the weathered samples had reached an estimated age of 2.6 years. It should be noted that only a narrow band of solar irradiation spectrum in the range 280–390 nm was considered as shown in Fig. 1, as the majority of material damage occurs within this range.

2.4. *Leaching experiment*

In order to simulate one-hour rainfall events, two rainfall intensities were selected for the leaching experiments: 5 mm/h (i1) and 15 mm/h (i2). The data for these intensities were obtained from the KNMI database [23]. The volume of water required to cover the 78 ×99 mm bio-composite sample surface for these intensities are 39 ml for i1 and 116 ml for i2.

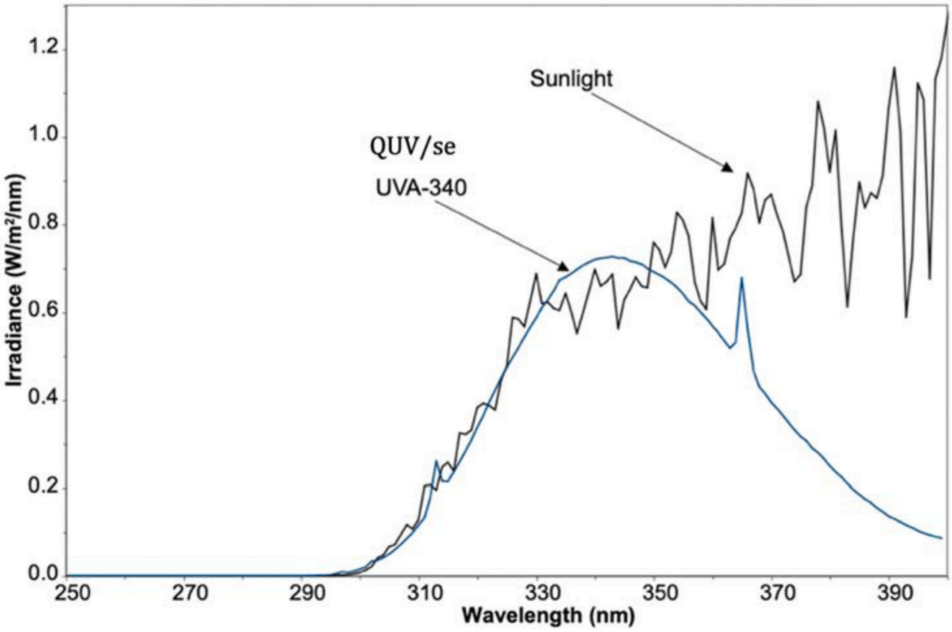


Fig. 1. Irradiation spectrum of the QUV/se weathering machine (blue line) and solar spectrum (black line) [26].

2.4.1. Rainwater simulation

The composition of the simulated rainwater, as detailed in Table 2 is based on the recommendations outlined by the European Commission [28].

The rainwater solution was stored for a period of seven days in order to enable stabilisation to occur, with measurements of pH taken on daily basis. The final pH was adjusted to the desired value of 6.5 with 1 M HCl (35 %) and 2 M NaOH solutions. Further details can be found in the Supplementary Material.

2.4.2. Experimental setup

Leaching test were conducted in order to assess the potential leaching of metals and resin compounds from novel bio-composite materials under precipitation exposures. Prior to the start of the experiment, all façade sample surfaces both new and weathered, were dusted, rinsed with the ultrapure water, and dried prior to the start of the experiment. Bio-composite samples of dimensions 78 ×99 mm were placed on a fixture inclined at 45°, as illustrated in Fig. 2, to ensure proper exposure of the surface to the simulated rainwater and adequate residence time of the droplets on the bio-composite surface [14]. A 45° angle was maintained by means of a supporting stand device for the façade sample. In order to prevent leaching from the support surface, an inert parafilm was applied and the support was then placed in a glass box. The rainwater was collected in the channel and placed underneath the sample. The façade sample surface was uniformly covered with the simulated rainwater using a multichannel pipette. The volume of each channel was 125 μL, with a total of eight channels used to deliver approximately 1.0 ml of water per load. The delivery of rain was conducted manually for a period of one hour, in order to simulate the duration of selected rainfall events at the specific rain intensity. In order to ascertain the requisite number of loads for the multichannel pipette, the volume necessary to cover the surface sample was divided by the volume of water delivered per load. For the first intensity (i1, 5 mm/h), 39 loads were required, resulting in the delivery of approximately 39 ml of water over the course of one hour. For the second intensity (i2, 15 mm/h), 116 loads were required, resulting in a total delivery of approximately 116 ml of water over the same period. The tests were performed in replicates - four for intensity i1 and two for intensity i2. Following the replicates, composite samples, which were mixtures of the effluent from each test, were collected, stored at 4 °C and sent for analysis. The composite samples were analysed by using ICP-MS method to detect the heavy metals. The samples were homogenised and acidified (HNO_3), after which the analysis was performed from the liquid phase. GC–MS was used to measure styrene in water samples. In the end, HPLC–UV was used to measure furfuryl alcohol in the composite samples.

It should be noted that the aforementioned leaching tests are not currently regulated by standards. However, these tests adhere to the principles set forth in the Dynamic Surface Leaching Test (DSLT), which is regulated by the European harmonized technical specification CEN/TS 16637–2 [29]. In regard to the horizontal exposure of the façade samples to rainfall, there are two scenarios: the terms "run-off" and "stagnation" are used to describe the movement of water. The 'run-off' scenario entails the rapid drainage of rainwater, whereas the 'stagnation' scenario (the standard Dynamic Surface Leaching Test) involves the rainwater remaining in contact with the samples for a longer duration. It is possible that construction elements which are exposed to intermittent wet-dry stress may exhibit a different leaching behaviour to those which are in permanent contact with water, with either an increased or a decreased release. In this study only the 'run-off' scenario has been selected to conduct leaching tests with the objective of evaluating the leaching after a one-hour rainfall event. It is important to note that the samples were initially tested for i1, then rinsed with deionized (DI) water and dried before being tested for i2 on the same samples.

2.4.3. Environmental risk assessment

The Environmental Risk Assessment (ERA) was carried out in accordance with established guidelines [20], the methodology, as detailed below. This assessment considered a real-case scenario regarding a pumping station with façade elements made from the bio-composite material, a receiving pond for the rainwater running off from the pumping station, and operating hours of the pumping station, as outlined in Section 2.4.4. The assessment accounted for a single rain event of duration 1 hour at intensity i1 (5 mm/h) and at intensity i2 (15 mm/h).

2.4.3.1. ERA Methodology. The Environmental Risk Assessment (ERA) conducted in this study exclusively considers the effects of chemical leaching from façade panels, which result in a specific concentration in the pond situated beneath. The background concentrations of the chemicals that are already present in the pond, as well as and the chemical composition of rainwater were not included in the study. The reasons for this are the absence of on-site data concerning the background concentrations of chemicals in the pond, and the utilization of rainwater composition recommendations established by the European Commission which lack

Table 2
Simulated rainwater composition.

Ion	Concentration [mg/L]
NH_4^+	0.910
Cl^-	1.960
Mg^{2+}	0.145
NO_3^-	2.010
PO_4^{3-}	1.000
SO_4^{2-}	1.460
pH	6.500

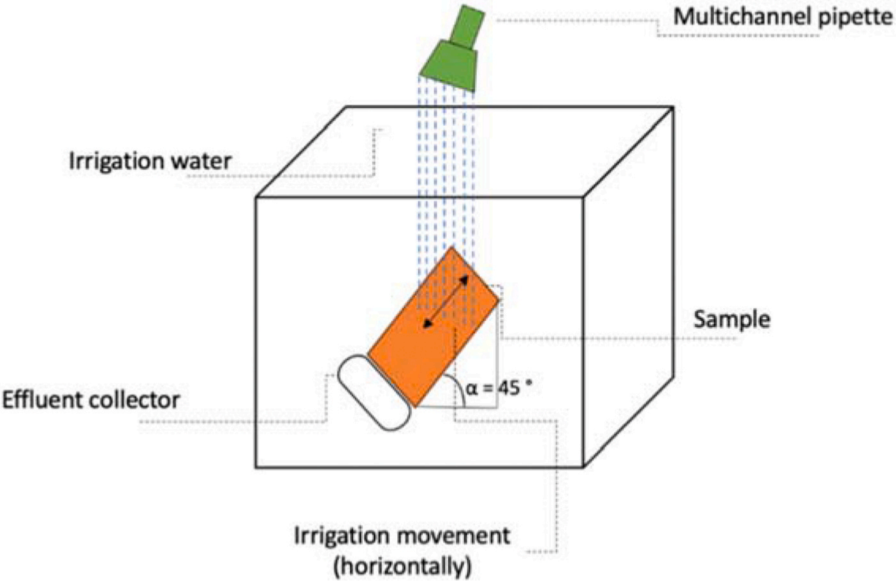


Fig. 2. Scheme of experimental setup of leaching tests using multichannel pipette.

geographical specificity of rainwater and its chemical composition in the Netherlands. When these two factors would have been taken into account, the results of the risk assessment may have differed. The leaching of chemicals, combined with the background concentrations that are already present in the pond and in the rainwater, could have resulted in the safety threshold being exceeded at an earlier stage.

To carry out the ERA, it is essential to follow the steps of the framework and define the risk objective, which in this case relates to the leaching of chemical such as heavy metals and resin compounds (VOCs) into surface water. The ERA methodology involves the following four main steps:

1. Hazard Identification: Presence of heavy metals and resin compounds in leaching effluent.
2. Dose-response model: Freshwater (where the façade elements considered in this study as real case scenario are placed in) is selected as the receiving ecosystem. The Predicted No Effect Concentrations (PNEC) for freshwater were obtained from European Chemical Agency (ECHA) database [22]. The PNEC represents the concentration of a substance below which adverse effects are unlikely to occur in both long-term and short-term exposure scenarios [22]. Table 3 shows the PNEC values of tested chemicals in freshwater in μg/l.
3. Exposure assessment: The Predicted Effects Concentration (PEC) is the predicted environmental concentration of the contaminant, which was calculated based on the leaching test results, as described in Section 2.4.4. The next step is to assess whether the pollutant can pose a threat at this concentration.
4. Risk characterization: In order to evaluate the actual environmental risk, based on the ERA guidelines [20], the environmental risk was calculated as follows:

$$Risk = \frac{PEC}{PNEC} \tag{2.1}$$

The PEC/PNEC ratio must be below the threshold of 1.00, for the risk to be considered acceptable [20,21].

Table 3
List of PNEC values for freshwater, from ECHA database [22].

Chemical [-]	PNEC [μg/L]
B	$2.9 \cdot 10^{3}$
Ba	$1.15 \cdot 10^{2}$
Cd	$1.90 \cdot 10^{-1}$
Cu	$6.3 \cdot 10^{0}$
Mg	$4.10 \cdot 10^{2}$
Mn	$3.40 \cdot 10^{1}$
Sb	$1.13 \cdot 10^{2}$
Zn	$1.44 \cdot 10^{1}$

2.4.4. Leaching test scenario

The facade elements examined in this study are applied to an existing pumping station in the Netherlands which has a façade made of bio-composite panels, as displayed in Fig. 3. the total surface area of the panels is 54 m^2 with a thickness of 6 mm. The pumping station is situated above a pond with dimensions of 12.5 m in width, 50 m in length and 1.25 m in depth. This pond is located within the Amsterdam Dune Area (Amsterdamse Waterleidingduinen), which is a managed aquifer recharge system that also functions as a nature reserve and protected environmental area of *Natura 2000* network [30].

The ERA was conducted by means of an examination of the leaching process from façade elements. Fig. 3 illustrates the vertical walls as a leaching surface for a single side of the construction. It should be noted that the roof is not constructed from bio-composite materials. The experimental leaching test was conducted to evaluate the impact of rain hitting a vertical wall at an angle of 45°. This angle was selected on the basis of the findings presented by Weiler and Vollpracht. [14], which indicated that this angle should result in the highest leaching and thus represents the worst-case scenario.

The present study was conducted with the objective of investigating the release of substances into surface water following a single rainfall event. The pump station is operational for only three days per year, resulting in the renewal of water three times per year. The prolonged stagnation of water, in the absence of a flow rate, serves to enhance the accumulation of heavy metals. However, due to the absence of consistent input data regarding leaching behaviour over time, this study considers solely the environmental impact of potential leaching following a single rainfall event from the bio-composite façade on one side of the pumping station building.

The potential environmental risk was evaluated using both deterministic and stochastic approaches, in order to gain a comprehensive understanding of the potential impact. The deterministic approach was employed to ascertain whether any of the chemicals exceeded the safety threshold. Subsequently, the stochastic approach was implemented in order to mitigate the limitations associated with the availability of some input data for the deterministic approach. Finally, a sensitivity analysis was conducted to gain a more comprehensive understanding of leaching behaviour by assessing the PEC/PNEC ratio in response to varying input data, including leaching concentrations and rainfall intensities. The sensitivity of the system was evaluated through the analysis of three distinct cases:

1. Sensitivity case 1 (s1): The objective of this sensitivity analysis was to assess the impact of varying leaching concentrations on the environmental risk (i.e. PEC/PNEC ratio). Furthermore, this sensitivity analysis was conducted to assess the reliability of the deterministic approach employed with limited input data. The samples collected from the leaching tests (representative of composite samples), were collected after four days (for rainfall intensity i1) and after two days (for rainfall intensity i2) of a wet-dry cycle. This was done in order to obtain a sufficient volume of effluent water for chemical analysis. Consequently, the observed leaching concentrations were interpreted as the cumulative leaching following one or more additional rainfall events. The cumulative leaching rate typically exhibits a rapid initial increase, followed by a constant rate [19]. Consequently, the leaching concentrations have been simulated in this sensitivity case using an exponential distribution.
2. Sensitivity case 2 (s2): The objective of this sensitivity case was to evaluate the impact of various rainfall intensities and to gain a deeper understanding of the leaching behaviour exhibited at varying rain intensities. A uniform distribution was employed to simulate rainfall intensities, with a minimum value of 5 mm/h (equivalent to rainfall intensity i1) and a maximum value of 15 mm/h (equivalent to rainfall intensity i2).

Fig. 3. Façade panels applied at the pumping station in a natural reserve [picture from D1.9 WIDER UPTAKE report].

3. Sensitivity case 3 (s3): In this sensitivity analysis, both leaching concentrations and rainfall intensities have been simulated simultaneously. This case study proved to be a valuable tool in identifying the primary critical parameters that influence leaching behaviour. Further details concerning the sensitivity analysis can be found in the Supplementary material.

To simulate a real-life scenario, the leaching test data were used to predict the potential release in the actual case. The concentration based on the surface area of the façade elements (the actual PEC value) was calculated using Eq. 2.2, as follows.

$$C_{L,real} = \frac{C_L * H_i * A_{1side}}{V_{pond}} \tag{2.2}$$

Where:

- $C_{L,real}$ [μg/l] represents the leaching concentration in the pond due to the leaching from the actual surface (A_{1side}) of façade elements into the pond with a volume V_{pond}. This value represents the Predicted Effects Concentration (PEC).
- C_L [μg/l] is the leaching concentration based on the leaching test results.
- H_i [m] is the height of the rain column (mm of rain in 1-hour duration converted in meter).
- A_{1side} [m^2] is the actual surface area of the one side of the vertical wall of pumping station construction.
- V_{pond} [m^3] is the volume of water in the pond estimated to be 781.250 m^3.

Two distinct scenarios were employed for the analysis of the environmental risk, each characterised by different precipitation intensities and a duration of one hour. The initial scenario evaluated the risk associated with a singular rainfall event, characterised by an intensity of 5 mm per hour (i1), while the subsequent scenario assessed the risk subsequent to a singular rainfall event, characterised by an intensity of 15 mm per hour (i2).

3. Results and Discussion

The weathering treatment leads to substantial change in the façade elements sample in terms of colour and texture of the surface, as displayed in Fig. 4.

Fig. 4 illustrates the two bio-composite materials M3 and M4, both new and weathered. The weathering treatment had an effect on the aesthetics of the materials, particularly in terms of colour and surface texture. The weathered samples, particularly M4, Fig. 4(d), exhibited a more irregular surface texture and a reduced water-resistant quality. The surface fibres, tightly pressed and glued together in fresh M4, were easily detached in weathered sample. The bio-composite samples before and after weathering were visually examined using an MBS-10 stereoscopic microscope equipped with an 8x eyepiece magnification and a 23 mm linear field of vision diameter. The selected objective magnification was 4x, resulting in a total magnification of 32x. This setup provided a field of vision in the object plane of 5.6 mm. The visual inspection of the weathered and not weathered samples using the MBS-10 stereoscopic microscope is presented in Fig. 5.

Looking at both Fig. 5 and Fig. 6, it is evident that both weathered samples exhibit alterations primarily in the surface colour. The surface texture seems slightly rougher for both weathered materials, with noticeable wrinkles visible at the microscopic imagine for M4, as displayed in Fig. 6. (d). In line with the aforementioned observations, it can be seen in Fig. 5(d) that the fibres are clearly distinguishable. Furthermore, the microscope image illustrated in Fig. 6(d) clearly indicated the presence of wrinkles, providing further evidence of weathering. The disparate weathering degradation of the two materials is likely attributable to their distinct composition and the different resin employed.

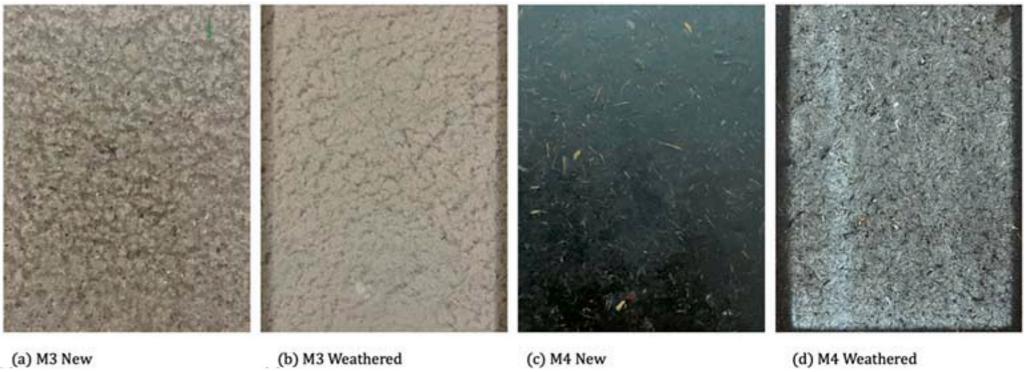
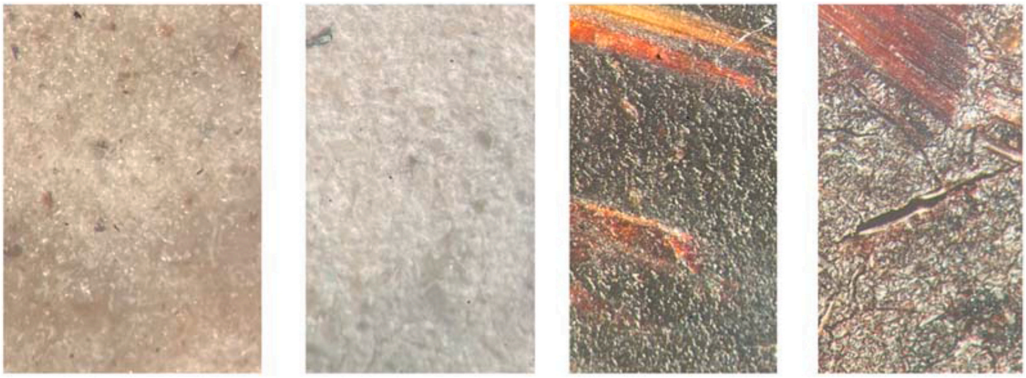

(a) M3 New (b) M3 Weathered (c) M4 New (d) M4 Weathered

Fig. 4. (a) Material M3 New; (b) Material M3 Weathered; (c) Material M4 New; (d) Material M4 Weathered.



(a) M3 New (b) M3 Weathered (c) M4 New (d) M4 Weathered

Fig. 5. MBS-10 Microscope images (32X magnification) of (a) Material M3 New; (b) Material M3 Weathered; (c) Material M4 New; (d) Material M4 Weathered.

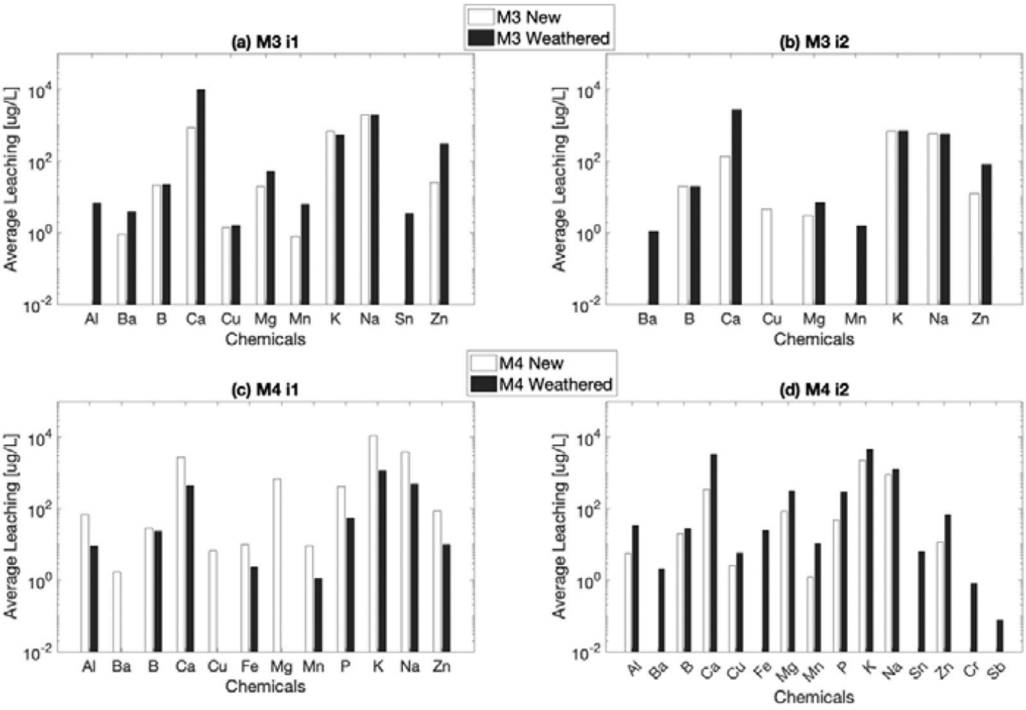


Fig. 6. Leaching test results after 1 hour of rain event, comparison between new and weathered samples: (a) M3 new and weathered samples at intensity i1 = 5 mm/h; (b) M3 new and weathered samples at intensity i2 = 15 mm/h; (c) M4 new and weathered samples at intensity i1 = 5 mm/h; (d) M4 new and weathered samples at intensity i2 = 15 mm/h.

3.1. Leaching test results

The results of the leaching test conducted on the bio-composite material samples, which were subjected to two precipitation intensities (i1 = 5 mm/h and i2 = 15 mm/h) for a duration of one hour, are presented in Fig. 7. It should be noted that leaching results represent actual leaching of the material, as background concentration of ions, added by simulated rainwater, was subtracted. The samples were subjected to analysis to ascertain the presence of heavy metals and for the presence of the resin compounds styrene and furfuryl alcohol. Neither styrene nor furfuryl alcohol was identified in the samples.

Fig. 7 illustrates the leaching behaviour of new samples in comparison to the leaching of weathered samples for each material, differentiated by the intensity of the precipitation under which they were tested. The results demonstrate clear differences in leaching patterns between materials M3 and M4, as well as between new and weathered samples, particularly for M4. With regard to M3 it can be observed that both the new and weathered samples display comparable leaching rates at precipitation intensities i1 (5 mm/h) and

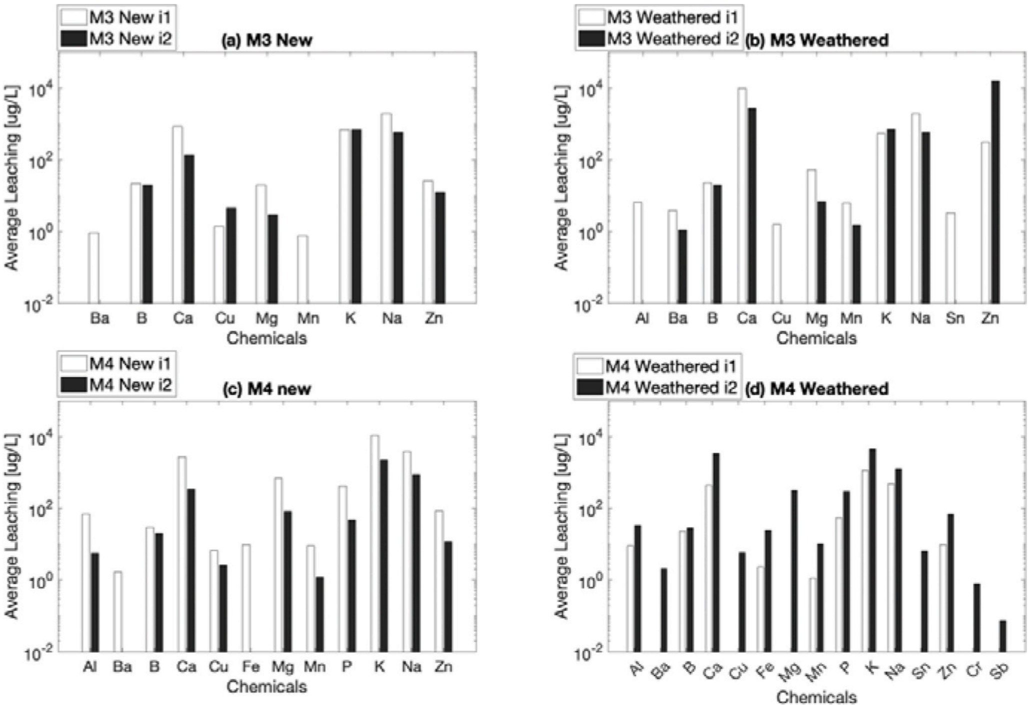


Fig. 7. Leaching test results, comparison between precipitation intensity i1 (5 mm/h) and precipitation intensity i2 (15 mm/h): (a) M3 new sample at both precipitation intensities; (b) M3 weathered sample at both precipitation intensities; (c) M4 new sample at both precipitation intensities; (d) M4 weathered sample at both precipitation intensities.

i2 (15 mm/h), with the exceptions of barium (Ba) at intensity i1, and calcium (Ca) and zinc (Zn) at both intensities. This is illustrated in Fig. 7(a) and Fig. 7(b). The elevated zinc release observed in M3 is likely attributable to the presence of zinc stearate, a release agent that resides on the surface, as previously noted in the footnote of Table 1. The weathered sample of M3 exhibits a discernible alteration in colour as depicted in Fig. 6(b) and more comprehensively in Fig. 7(b). This exposure is likely to increase water penetration, a phenomenon that was also observed during the course of the testing. Consequently, this results in an increase in the leaching of water-soluble chemicals.

With regard to M4, as illustrated in Fig. 7(c, d), there is a noticeable discrepancy in leaching behaviour between the new and the weathered samples. Specifically, at rain intensity i1 (5 mm/h), the new sample exhibits greater leaching compared to the weathered sample. Conversely, at rain intensity i2 (15 mm/h), the weathered sample demonstrates increased leaching relative to the new sample. Notably, the leaching of metals such as chromium (Cr), iron (Fe), tin (Sn) and antimony (Sb) is exclusive to the weathered sample of M4. This observation implies that the weathering process may have inflicted more significant surface degradation on M4 than on M3. Surface degradation can be seen in Fig. 6(c, d), and in more details in Fig. 6(c, d).

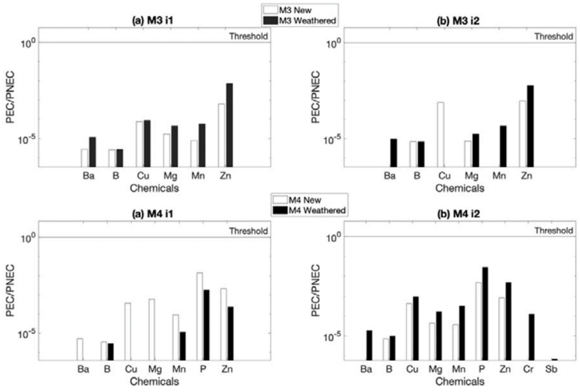


Fig. 8. Environmental Risk Assessment for M3: (a) M3 new and weathered material at precipitation i1 (5 mm/h) for one single rain event; (b) M3 new and weathered material at precipitation i2 (15 mm/h) for one single rain event; (c) M4 new and weathered material at precipitation i1 (5 mm/h) for one single rain event; (d) M4 new and weathered material at precipitation i2 (15 mm/h) for one single rain event.

The alteration in the surface texture of the weathered M4 sample reveals the fibres within the bio-composite material, which, as previously observed for M3, increases water penetration, and activates the leaching of heavy metals present in the material at lower levels. Additionally, findings of Quero, Ballesteros [24] indicated that olive pamoate, which is employed as a release agent for this material, may be contaminated with metals such as boron (B), calcium (Ca), potassium (K), magnesium (Mg), sodium (Na), manganese (Mn), and zinc (Zn). As with the results observed for material M3, where the elevated release of Zn was linked to the use of zinc stearate as a release agent, the significant leaching of Ca, K, Mg, Na, Mn, and Zn in the weathered samples of material M4 can likely be related to the use of olive pamoate as a release agent.

A further comparison can be made between the bio-composite samples tested under different precipitation intensities. It is important to note that the samples were first tested for i1 (5 mm/h), then rinsed with ultra-pure water and dried before being tested for i2 (15 mm/h) on the same samples. It is possible that the aforementioned sequence of actions may have had an impact on the leaching results at intensity i2. However, the impact of this is uncertain, given the observed variations in leaching patterns between both M3 and M4, as well as between new and weathered materials. Fig. 8 displays a comparison for two rain intensities analysed.

Fig. 8 illustrates the variations in leaching patterns under subsequent precipitation intensities: intensity i1 (5 mm/h) and intensity i2 (15 mm/h) for the new and weathered bio-composite façade samples M3 and M4. As in the previous case (Fig. 7), a notable distinction in leaching behaviours between M3 and M4 can be observed.

Fig. 8(a, b) illustrate the leaching concentrations of new and weathered samples of M3, respectively. In both cases, higher leaching concentrations are observed at intensity i1, except for Zn in the weathered sample, which may be due to the zinc stearate used as agent release, as mentioned above. This pattern can be explained by the dilution effect observed during an hour of rainfall intensity i2, where a larger volume of water dilutes the concentration of leached substances. Furthermore, as previously stated, the samples were initially tested for i1, and then, following rinsing with ultra-pure water and drying, the same sample was tested for i2. This sequence of actions may have affected the leaching results at intensity i2.

The leaching of zinc is more pronounced in the weathered sample in the presence of higher precipitation intensity, indicating the potential for internal accumulation of zinc beyond surface deposition from the release agent. This behaviour indicates that in weathered samples, zinc is more easily mobilised under higher water volumes, highlighting the impact of rainfall intensity on leaching dynamics.

Regarding material M3, a completely opposite pattern is noticeable between the new and weathered materials. Fig. 8(c) shows that the highest leaching concentrations occur after one hour of precipitation at rain intensity of 5 mm/h (i1), which aligns the observation made for M3 about the dilution effects and the sequence of the experiments followed, resulting in lower leaching concentrations associated with the rain intensity of 15 mm/h (i2). Turning to Fig. 8(d), the weathered sample exhibits the opposite behaviour in terms of leaching, resulting in higher concentrations after intensity i2. This includes the leaching of chemicals which are only observed in the weathered sample under these conditions. This is in line with the previous consideration that the altering of the surface of the weathered M4 sample, which changes both its colour and texture, exposes the fibres, and increases leaching, particularly under higher precipitation intensity.

The percentage leaching release was calculated based on the total chemical mass of substances leached from materials M3 and M4 and chemical mass of these substances in the bio-composite materials. The chemical analysis of the raw ingredients was previously described by Nativio, O. [6] and additional details can be found in the Supplemental Material. The calculated percentage leaching release for M3 and M4, both of which were designated as "New" (N) and "Weathered" (W) samples, are presented in Table 4 and Table 5, respectively.

The data presented in Table 5 indicate a notable release of B (~ 31 %) and Zn (~ 10 %), for the weathered sample of M3, following various precipitation events at both rain intensities (i1 = 5 mm/h and i2 = 15 mm/h). Similarly, Table 2 demonstrates that for M4, the leaching of K from the weathered sample can reach the maximum approximately 68.3 % after two precipitation events at rain intensity of 15 mm/h (i2). Furthermore, the leaching percentages of B in both the new and weathered samples fall within the range of 10–32 % for both precipitation intensities. The pronounced leaching observed is related to the water solubility of these chemicals, which allows their release during the one-hour precipitation event mentioned above. Although long-term leaching trend data are lacking, the findings of the previous study by Nativio, O. [19] suggest that the leaching of substances initially found at elevated concentrations is likely to decrease over time. This decrease is due to the gradual depletion of the leachable chemical reservoir within the material,

Table 4
Percentage leaching releases based on the initial chemical contamination of M3.

% leaching	Rainfall intensity i1		Rainfall intensity i2	
	M1 N	**M1 W**	**M1 N**	**M1 W**
Al	0.00	$6.00 \cdot 10^{-2}$	0.00	0.00
Ba	$9 \cdot 10^{-2}$	$4.00 \cdot 10^{-1}$	0.00	$3.40 \cdot 10^{-1}$
B	$1.16 \cdot 10^{1}$	$1.22 \cdot 10^{1}$	$3.16 \cdot 10^{1}$	$3.16 \cdot 10^{1}$
Ca	0.00	$1.00 \cdot 10^{-2}$	0.00	$1.00 \cdot 10^{-2}$
Mg	0.00	$1.00 \cdot 10^{-2}$	0.00	0.00
Mn	$1.00 \cdot 10^{-2}$	$5.00 \cdot 10^{-2}$	0.00	$4.00 \cdot 10^{-2}$
P	0.00	0.00	0.00	0.00
K	$4.80 \cdot 10^{-1}$	$3.90 \cdot 10^{-1}$	$1.42 \cdot 10^{0}$	$1.44 \cdot 10^{0}$
Na	$2.10 \cdot 10^{0}$	$2.11 \cdot 10^{0}$	$1.84 \cdot 10^{0}$	$1.84 \cdot 10^{0}$
Zn	$9.80 \cdot 10^{-1}$	$1.17 \cdot 10^{1}$	$1.43 \cdot 10^{0}$	$9.21 \cdot 10^{0}$

Table 5
Percentage leaching releases based on the initial chemical contamination of M4.

% leaching	Rainfall intensity i1		Rainfall intensity i2	
	M2 N	M2 W	M2 N	M2 W
Al	$9.00 \cdot 10^{-2}$	$1.00 \cdot 10^{-2}$	$2.00 \cdot 10^{-2}$	$1.40 \cdot 10^{-1}$
Ba	$1.00 \cdot 10^{-2}$	0.00	0.00	$2.00 \cdot 10^{-2}$
B	$9.80 \cdot 10^{0}$	$8.59 \cdot 10^{0}$	$2.01 \cdot 10^{1}$	$3.11 \cdot 10^{1}$
Ca	0.00	0.00	0.00	$1.00 \cdot 10^{-2}$
Cu	$1.00 \cdot 10^{-1}$	0.00	$1.10 \cdot 10^{-1}$	$2.80 \cdot 10^{-1}$
Fe	0.00	0.00	0.00	$3.00 \cdot 10^{-2}$
Mg	$1.10 \cdot 10^{-1}$	0.00	$1.10 \cdot 10^{-1}$	$1.70 \cdot 10^{-1}$
Mn	$1.40 \cdot 10^{-1}$	$2.00 \cdot 10^{-2}$	$6.00 \cdot 10^{-2}$	$5.50 \cdot 10^{-1}$
P	$1.40 \cdot 10^{-1}$	$2.00 \cdot 10^{-2}$	$3.70 \cdot 10^{-1}$	$3.20 \cdot 10^{-1}$
K	$5.01 \cdot 10^{1}$	$5.69 \cdot 10^{0}$	$4.67 \cdot 10^{1}$	$6.83 \cdot 10^{1}$
Na	$1.53 \cdot 10^{0}$	$2.10 \cdot 10^{-1}$	$18.15 \cdot 10^{0}$	$1.66 \cdot 10^{0}$
Sn	0.00	0.00	0.00	$6.64 \cdot 10^{0}$
Zn	$5.20 \cdot 10^{-1}$	$7.00 \cdot 10^{-2}$	$2.10 \cdot 10^{-1}$	$1.36 \cdot 10^{0}$
Cr	0.00	0.00	0.00	$4.50 \cdot 10^{-1}$

leading to a stabilisation of leaching patterns over time.

A comparison of the percentage leaching for both M3 and M4 (Table 4 and Table 4) with the leaching results (Fig. 8, as well as Fig. 8) reveals a discrepancy in calcium leaching for both new and weathered façade samples for both M3 and M4, respectively. It can be reasonably concluded that most of the calcium leaching cannot be attributed to the original contamination of the raw materials. It is likely that a portion of the calcium leaching originated from other contaminants, such as additives and resins, which were not verified for this study.

3.2. *Environmental risk assessment*

The Environmental Risk Assessment evaluated the impact of leaching from new and weathered materials (both M3 and M4) for rain intensities of 5 mm/h (i1) and 15 mm/h (i2). The assessment aimed to determine whether the leached concentrations exceeded the PEC/PNEC threshold of 1.00 [20]. The evaluation of the environmental risk assessment has been done by considering only the chemicals for which the PNEC values are available in European Environmental Agency database [22]. Thus, chemicals such as Al, Ca, Fe, K, Sn, and Na were not considered in the assessment. The obtained results are shown in Fig. 9.

As illustrated in Fig. 9, the environmental risk assessment demonstrated that the risk levels for both materials M3 and M4 remained below the safety threshold of 1.00 for both rain intensities.

The leaching tests conducted in the laboratory on the M3 and M4 bio-composites revealed significant leaching of certain components, as shown in Fig. 7 and Fig. 8. However, even at these high levels of leaching, the results remained below established thresholds for both new and weathered samples under a range of environmental conditions, including normal (i1) and high (i2) rainfall

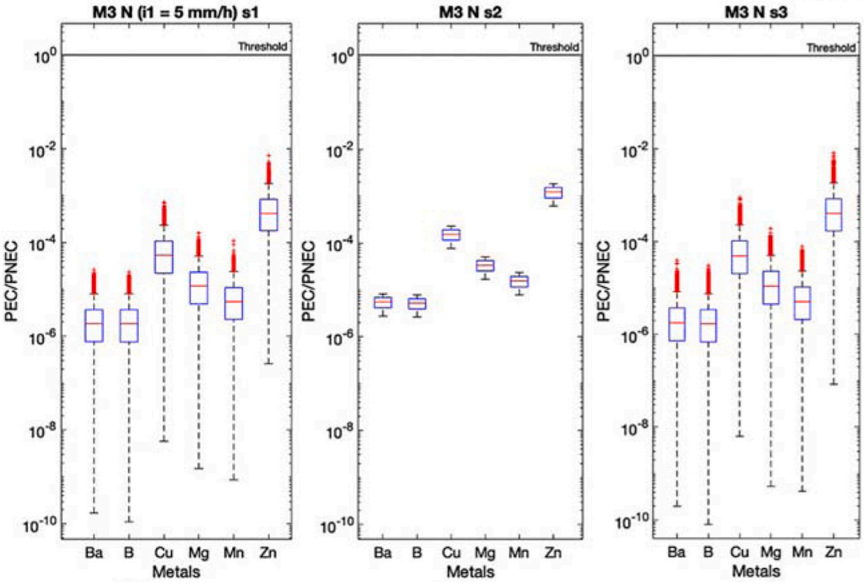


Fig. 9. Comparison sensitivity cases for M3 New sample: (a) sensitivity case s1; (b) sensitivity case s2; (c) sensitivity case s3.

intensities. This consistency across different test conditions suggests that leaching does not pose a risk of exceeding safety or environmental standards. Therefore, it can be stated that the use of these bio-composite materials as façade elements is acceptable from the environmental risk perspective. These results support the suitability of M3 and M4 bio-composites for exterior applications, ensuring compliance with safety regulations and maintaining environmental integrity. However, background concentrations of both rainwater and surface water need to be considered for a comprehensive risk assessment.

The limitations of an environmental risk assessment based solely on laboratory data were addressed by employing a stochastic approach. In this approach, variables such as leaching concentration and rainfall intensity were treated as uncertain. The objective was to analyze the potential impact of realistic variations in these values, which could occur in real-world conditions. The objective was not only to ascertain whether such variations could elevate the overall environmental risk levels potentially increasing the PEC/PNEC values above the threshold of 1.00, but also to gain insight into the leaching behavior under varying input parameters.

The stochastic approach is based on the Monte Carlo method with 10,000 trials. The selection of 10,000 trials was based on the observation that no significant changes were observed when the number of iterations was increased beyond this value (analysis not shown here). Two sensitivity cases were modelled, s1 with uncertain leaching concentrations, and s2 with uncertain rainfall intensity, both using pre-specified probability density functions, as described in Section 2.4.4.

Fig. 8 and Fig. 10 illustrate the influence of the aforementioned input parameters on the PEC/PNEC ratio for materials M3 and M4, respectively. The results presented in this study pertain solely to the “new” samples at fixed rain intensity i1 = 5 mm/h for sensitivity case s1.

As illustrated in Fig. 10 and Fig. 11, there was no significant increase in the PEC/PNEC ratios and the values remained below the safety threshold for all heavy metals and across all three sensitivity cases.

A comparison of Fig. 7(a) and Fig. 8(b) for M3, as well as Fig. 7(a) and Fig. 8(b) for M4, reveals that the sensitivity s1 case is distinguished by a wider boxplot in comparison to the sensitivity s2 case. Furthermore, the impact of varying the leaching concentrations is evident in the sensitivity s3 case (Fig. 6(c) and Fig. 7(c) for M3 and M4 respectively), which also exhibits a distinguishably wider boxplot shape. This suggests that the leaching concentration is the parameter that has the greatest impact on the PEC/PNEC ratio, rather than rainfall intensity.

A further significant outcome of this sensitivity analysis was the observation that, in this specific case study, the deterministic approach, due to the limited availability of input data (including on-site data that is lacking, as well as composite samples representing leaching under various rainfall intensities and composite samples representing leaching behavior over time), tends to overestimate the overall risk. This can be seen by comparing the deterministic results to the stochastic (i.e. Monte Carlo simulation) results for sensitivity case s1, as shown in Fig. 8 for material M4 at intensity i1 (new sample).

As illustrated in Fig. 8, the boxplot represents the outcomes obtained from sensitivity s1 case for M4 at intensity i1. The scatter points, in contrast, are representative of the deterministic results. As shown, the scatter points exceed the boundaries of the boxplot, indicating that the deterministic approach tends to overestimate the overall PEC/PNEC ratio. To confirm this, the probability density function (PDF) used to simulate the leaching concentrations was analyzed by comparing the average values obtained from the sensitivity analysis with the results obtained from the deterministic approach. The comparative results are presented in Fig. 12.

Fig. 12 shows the histogram of the simulated PEC/PNEC ratios based on the exponential probability density function used for simulating leaching concentrations. The y-axis represents the probability density, which indicates the frequency with which different PEC/PNEC ratios (represented on x-axis) are likely to occur according to the sensitivity analysis results. The vertical blue dashed line represents the average PEC/PNEC ratio values obtained from the sensitivity analysis, while the red vertical line represents the deterministic PEC/PNEC ratio results. As it can be seen from the plots, the deterministic values yield a higher PEC/PNEC ratio, which is characterized by a lower frequency of occurrence, in comparison to the values obtained from the sensitivity analysis. This observation is consistent with the findings presented in Fig. 10.

A comparison between the deterministic results and sensitivity s2 case has been conducted, revealing a significantly different outcomes from those of the earlier comparison between the deterministic approach and sensitivity case s1. Upon varying the rainfall intensities, the average values estimated by the Monte Carlo analysis and the deterministic approach exhibited minimal discrepancy, as illustrated in Fig. 13. Further examination of Fig. 13 reveals a histogram representative of the uniform distribution employed to simulate the rainfall analysis. The vertical lines representing the Monte Carlo average value (dashed blue vertical line) and the deterministic results (red vertical line) are in close proximity and frequently overlap. This demonstrates that variability in rain intensity has a predictable influence on the PEC/PNEC ratios, effectively captured by the deterministic approach. Overall, the environmental risk (PEC/PNEC) is more sensitive to variations in leaching concentration than to variation in rainfall intensity. The deterministic approach overestimates risk when leaching concentration varies (as stated above) but aligns with the stochastic approach when rainfall intensity varies. In conclusion, efforts to mitigate environmental risk should focus more on controlling leaching concentrations rather than rainfall intensities.

In the view of these findings, it would be both significant and interesting to replicate the laboratory leaching tests by evaluating more than two rainfall intensities and assessing the potential leaching over time. This would facilitate the identification of the distribution that most closely aligns with the cumulative leaching release for each chemical. In accordance with the aforementioned, it is of considerable significance to gather on-site data with the objective of validating the results of the laboratory tests and to gain a deeper comprehension of the leaching behavior in a real-world context.

4. Conclusion

This study presents a methodology for the assessment of environmental risks associated with the potential leaching of bio-

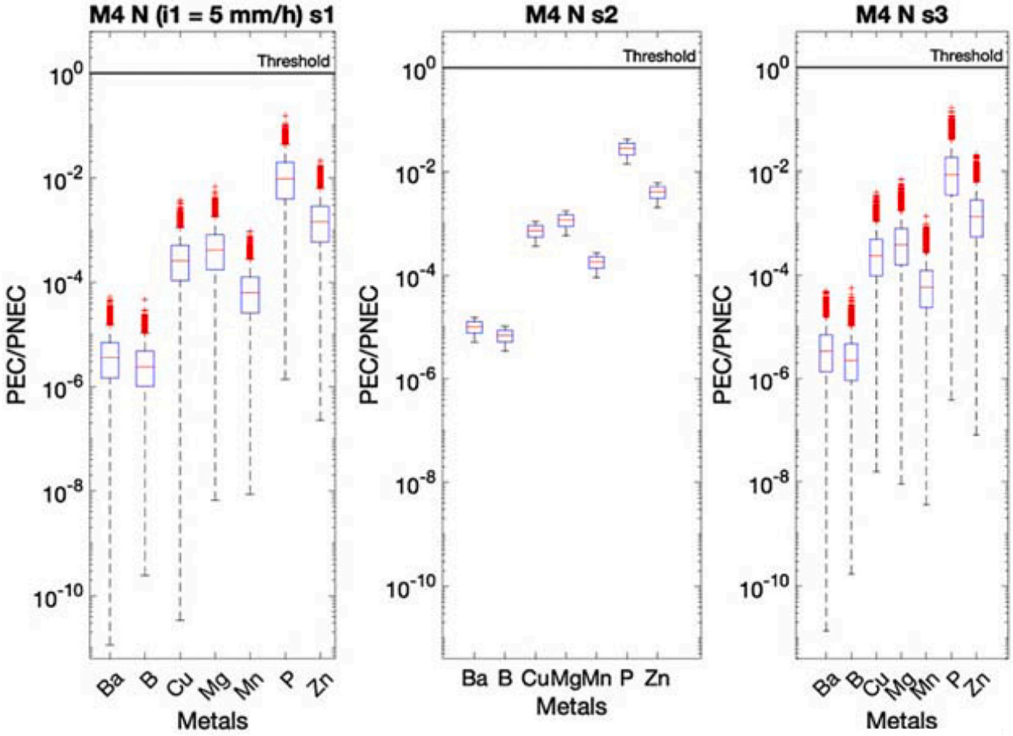


Fig. 10. Comparison sensitivity cases for M4 New sample: (a) sensitivity case s1; (b) sensitivity case s2; (c) sensitivity case s3.

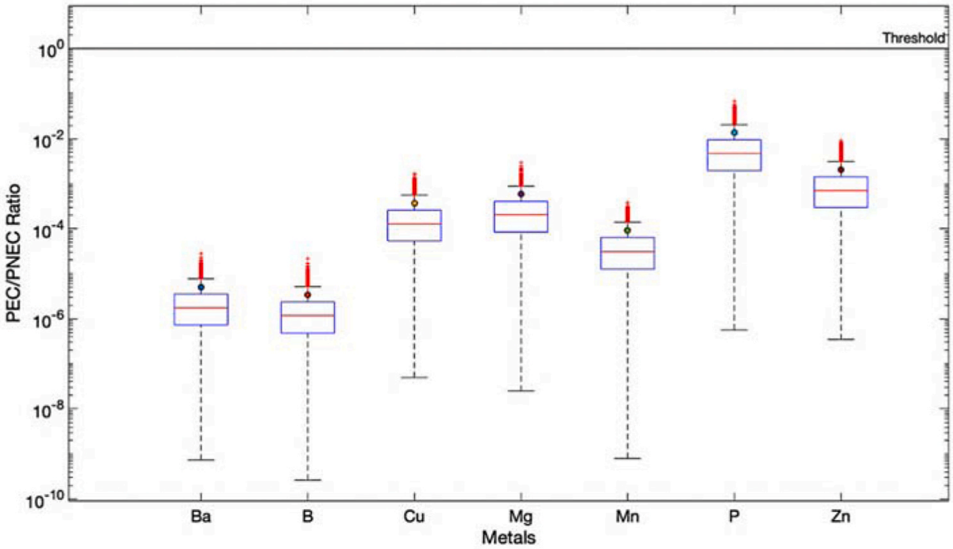


Fig. 11. Comparison deterministic approach and sensitivity case s1 Material M4 - New sample at intensity i1 (5 mm/h).

composite materials-based building façade panels into surface water. The façade panels were made of two new bio-composite materials (M3 and M4). Each material was tested in its new and weathered form, the latter being subjected to cyclic UV irradiation and high humidity. In addition, laboratory based leaching tests using simulated rainfall were conducted to assess the amount of heavy metals, resins and other potentially harmful substances leaching back into the natural aquatic environment. Leaching tests were performed using two different rainfall intensities analysed, everyday rainfall of 5 mm/h (i1) and the more extreme rainfall of 15 mm/h (i2). The data generated by laboratory leaching tests was, at the end, used to perform the environmental risk analysis for a real-life pumping station situation in the Netherlands, located in a nature reserve. The existing European ERA framework [20] was used for the risk

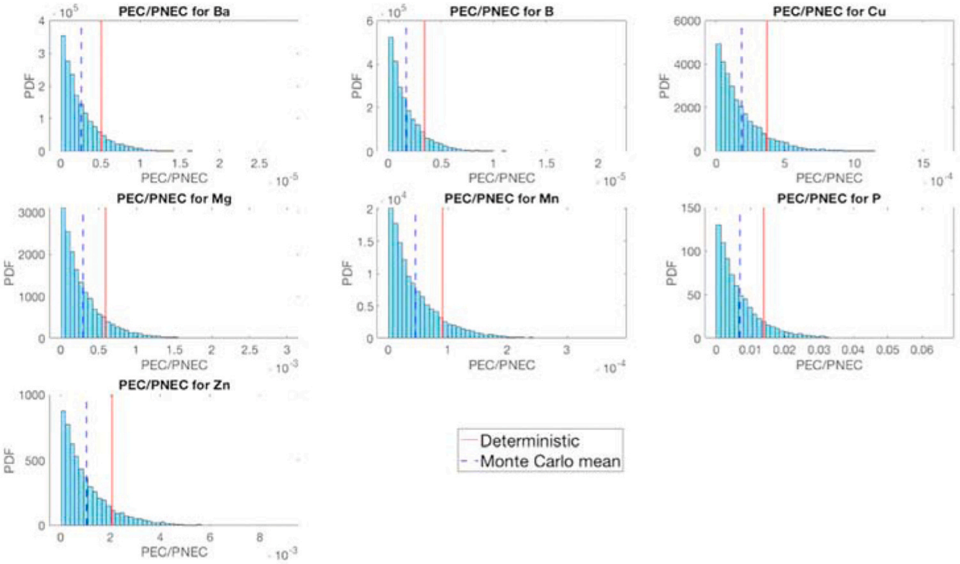


Fig. 12. Comparison between average values obtained from sensitivity case s1 analysis and the deterministic approach results, for material M4 “New” at i1 = 5 mm/h.

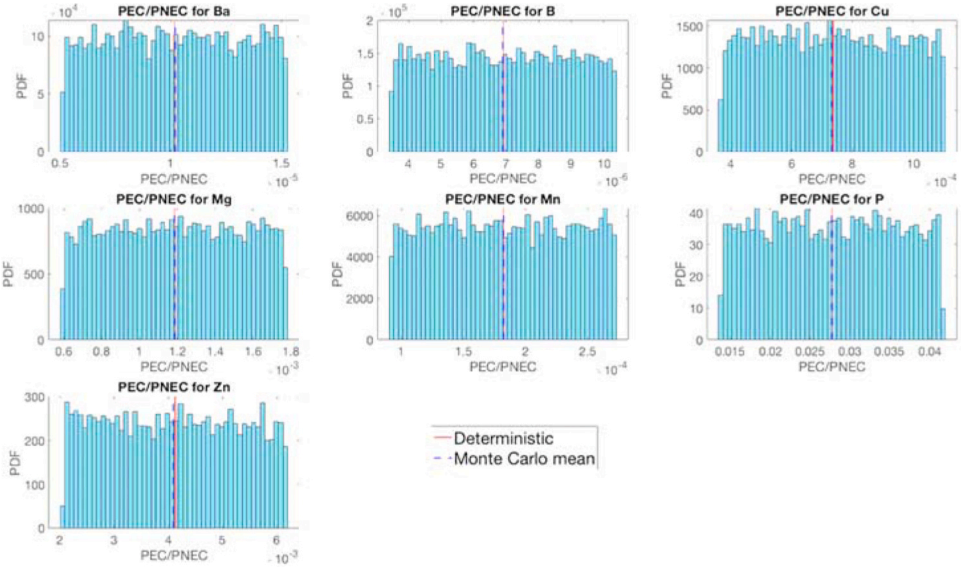


Fig. 13. Comparison between average values obtained from sensitivity case s2 analysis and the deterministic approach results, for material M4 “New”.

assessment. The key novelty of this work is in (a) the generation of new data by conducting laboratory leaching tests described above, and (b) the utilization of this data to perform the first environmental risk assessment involving new bio-composite materials. This represents a significant advancement as the corresponding field data currently does not exist. The approach presented here offers a practical, preliminary solution until such data becomes available.

Based on the results obtained following key findings are reported:

1. Regarding the environmental risk assessment, the results obtained showed that the risk levels associated with potential leaching from the two analysed bio-composite materials M3 and M4, both for new and weathered samples and for two rainfall intensities analysed, remained below the safety threshold, as defined by the European Risk Assessment framework.
2. The deterministic approach was found to be overly conservative in the risk assessment for this particular bio-composite application, when leaching concentrations were analyzed stochastically (sensitivity case s1), resulting in a higher PEC/PNEC ratio characterized by a lower frequency of occurrence based on the sensitivity analysis results.

3. The leaching concentration, as determined by the sensitivity analysis, appears to have the greatest influence on the PEC/PNEC ratio, greater than the rainfall intensity. Indeed, the variability in rain intensity does not significantly contribute to the uncertainty in the PEC/PNEC ratios. This is also in accordance with the observed results of the leaching tests, which indicated a lower leaching concentration at higher rainfall intensities. This was observed to be the case for all bio-composite materials tested except for material M4 weathered samples, which exhibited a significantly altered leaching behavior due to the impact of the weathering treatment, which led to a significative surface degradation.
4. The leaching test results obtained for material M3 showed that the leaching concentrations between new and weathered samples were generally comparable, with notable exceptions being calcium (Ca) and zinc (Zn) for both tested rain intensities. Interestingly, M3 displayed a higher leaching concentration at a lower precipitation intensity (5 mm/h) compared to the higher intensity (15 mm/h), with zinc (Zn) exhibiting increased leaching at the higher intensity. The highest leaching percentage observed for material M3 was approximately 31 % for boron (B) at rain intensity 15 mm/h, affecting both new and weathered materials.
5. The leaching test results obtained for material M4 indicated a higher leaching from the new sample compared to the weathered sample at intensity 5 mm/h. This trend was reversed at intensity 15 mm/h, where the weathered sample of M4 demonstrated a greater leaching effect, including the leaching of chromium (Cr), tin (Sn), and antimony (Sb) exclusively under these conditions. The leaching of barium (Ba), potassium (K), and sodium (Na) was also significant when assessed against the initial chemical concentrations in M4. Furthermore, the leaching rate of potassium (K) for material M4 reached a peak of approximately 69 %, indicating a higher propensity for leaching. This increased leaching could be linked to the release agent employed, which has not undergone chemical analysis, only the raw materials has been tested.
6. The effect of the weathering treatment varied considerably between the different materials, mainly due to their composition, particularly the type of resin used. For example, material M3, which is made of polyester resin, exhibited aesthetic changes because of weathering, particularly in terms of colour and surface texture. In contrast, the weathered sample of material M4, made of furan resin, showed increased roughness, and reduced water resistance. Originally, the surface fibres of new M4 sample were tightly compressed and firmly bonded, but in the weathered sample these fibres tended to detach. In addition, microscopic examination (Fig. 3), revealed significant wrinkling on the weathered M4, further illustrating the significant effects of the weathering process.

For future research, it is essential to extend the leaching tests over time. This would facilitate a more comprehensive understanding of long-term leaching behaviours, thereby enabling the generation of more consistent data for statistical analysis and the improvement of environmental risk assessment. Moreover, conducting field leaching tests would be of great significance in validating the consistency of laboratory findings under real environmental conditions, thereby enhancing the reliability of lab-to-field predictions. Furthermore, additional research is necessary to investigate the potential for leaching into the soil and groundwater, as this remains an unexplored pathway for possible pollution from using new bio-composite materials. In conclusion, the implementation of this methodology in other bio-composite applications and industries, such as infrastructure or packaging, would serve to enhance the impact of this approach, thereby ensuring sustainable practices across a range of sectors.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests. Zoran Kapelan reports financial support was provided by Horizon Europe - WIDER UPTAKE Grant n. 869283. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.cscm.2024.e03908.

Data Availability

The data that has been used is confidential.

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