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Investigating the ecotoxicity of construction product eluates as multicomponent mixtures

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Abstract

Background The release of hazardous compounds from construction products can harm human health and the environment. To improve the sustainability of construction materials, the leaching of substances from construction products and their potential environmental impact should be assessed. Twenty-seven construction products from different product groups were examined with a combination of standardized leaching tests (dynamic surface leaching test and percolation test) and biotests (algae, daphnia, fish egg, luminescent bacteria, umu and Ames fluctuation tests). To identify the released substances, extensive qualitative and quantitative chemical analyses were performed, including gas chromatographic and liquid chromatographic screening techniques.

Results Many of the tested eluates caused significant ecotoxic effects. Particularly high ecotoxicities were observed for grouts (lowest ineffective dilution (LID) up to 16384) and cork granules (LID up to 24578). The results of ecotoxicity tests allow the prioritization of the eluates that should be subjected to detailed chemical analyses. Organic screening by different methods and ranking the identified substances based on recorded hazard classification is a suitable approach to identify the relevant toxic substances.

Conclusions Determining the ecotoxicity of eluates from construction products records the summary effect of all leachable substances. This instrument is especially useful for construction products of complex and largely unknown composition. The ecotoxicological and the chemical–analytical approach complement each other in an ideal way to characterize the potential hazard of eluates from construction products and to identify the environmentally hazardous components in these eluates. Our results confirm that the proposed harmonized methods for testing eluate toxicity are an adequate and applicable procedure to move toward a more sustainable way of building and to reduce toxic effects of construction products in their use phase in the environment.

Keywords Construction products, Building materials, Ecotoxicity, Biotests, Leaching, Eluates, Chemical analysis, Analytical screening techniques, Mixture toxicity

Introduction

The building industry is an important sector worldwide and has a large environmental impact. Most life cycle assessments focus on the extensive CO₂ emission and the energy and material consumption in the building sector [1, 2]. Another environmental impact that should not be overlooked is the release of hazardous compounds from construction products that can harm human health and the environment. The materials used for the construction of buildings and infrastructure are complex mixtures with often unknown or only partially known composition

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[3]. They often contain many different chemicals, including organic additives with potentially hazardous properties. For many of these substances, the (eco)toxicological potential is not known and specific analytical methods and environmental standards are lacking.

During the phase of construction and service life, hazardous substances can be leached from construction products through contact with water, i.e., during rain events or when they are in contact with seepage or groundwater. The leaching of substances from construction products has been shown for various materials, such as façades [4–6], roof membranes [7], paints [8], bituminous sheets [9, 10], concrete [11–13], and cement [14]. Basically, all construction products that are used for the envelope of buildings (roof, façade, foundation), for the design of outdoor surfaces (e.g., terraces, sidewalks, fences, sport fields), or for the infrastructure (e.g., sewage pipes, insulation materials for pipelines) may risk the release of potentially hazardous substances. Another way that harmful substances can enter the environment is through emissions into the air. This emission pathway is particularly important for building products used indoors. For pollutants entering indoor air, mainly toxic effects on humans are considered. In the outdoor environment leaching is the predominant route of hazardous substances used in building materials raising concern. In addition, microplastics used, e.g., when constructing sports and recreational facilities are currently discussed as contaminants of emerging concern.

Up to now, new construction products are developed without investigating if substances can be leached from the product and if they may have adverse effects in the environment. To improve the sustainability of construction materials, it is thus desirable to assess the leaching of substances from construction products and their potential environmental impact before placing them on the market. In the EU, the regulations on construction products acknowledge that health and environmental aspects need to be considered over the entire life cycle of the products [15]. Furthermore, the European Commission has recently set the target of a toxic-free environment with the EU Zero Pollution Action Plan [16], including the aim of less pollution from, in, and around buildings. The EU's chemical strategy for sustainability aims to boost innovation for safe and sustainable chemicals to better protect humans and the environment [17]. The chemical strategy also seeks to assess the mixed toxicity effect of chemicals. This "cocktail" or sum effect of many different chemicals is also relevant for the assessment of construction products.

To reliably assess the emission of hazardous substances from construction products, harmonized test methods are a prerequisite. The standardized

leaching test methods were developed in the last decade by the Technical Committee CEN/TC 351 in the series "Construction products—Assessment of release of dangerous substances" prEN 16637:2021 part 1 to part 3 [18–20]. The horizontal dynamic surface leaching test (part 2, 'DSLIT' [19]) and the horizontal up-flow percolation test (part 3 [20]), were developed as standards for monolithic construction products and for granular construction products, respectively. These methods' robustness of applicability and sensitivity to test parameters were investigated for several construction products [21].

The eluates obtained in the leaching tests can be analyzed by means of substance-specific analysis for compounds that are known to be toxic when exceeding defined threshold values. However, this approach has its limits, since, as outlined above, construction products often contain various organic substances for which no specific analytical methods are available and for whose hazard potential data are scarce or missing. An alternative approach is to combine leaching tests with ecotoxicity tests. Here, the summary effect of all leached substances is recorded without analytical effort. The robustness of this approach was verified in two round robin tests [22–24]. In the European standardization process, the technical report CEN/TR 17105 [25] on the ecotoxicological testing of construction products was very recently updated into a technical specification CEN/TS 17459 [26], giving it a normative status. This progress in standardization is important, since the assessment methods need to be harmonized to make results comparable. Once information on the emission of hazardous substances is available, builders and consumers have the possibility to choose construction products that are ranked as environmentally friendly within the product group.

Data obtained by the described standardized methods on the ecotoxicological effects of substances leached from construction products are still scarce. Therefore, this study investigated several construction products from different product groups that are used for the envelope of buildings or other exterior surfaces. The first objective of this study was to demonstrate the applicability of the ecotoxicity tests listed in CEN/TS 17459 [26]. Based on these data, conclusions can be drawn on what kinds of products are particularly critical and whether the emission performance varies greatly among the products of one product group. If this is the case, construction products that are less critical could be promoted, for instance by establishing an eco-label for this product group or defining new regulatory requirements in the future. The use of ecotoxicity tests for this purpose is in line with the recommendation to integrate effect-based methods for the diagnosis and monitoring of water quality [27].

The second main task of this study was to investigate the “black box “ of organic substances leached from construction products by different organic screening methods. It was attempted to link the observed ecotoxicity in the eluates with specific chemical compounds in the complex mixture of construction product eluates. Overall, this study shows that the combination of leaching and ecotoxicity tests is a valuable tool for assessing hazardous substances leached from construction products.

Materials and methods

Products

In total, 27 construction products from eight product groups were examined (Table 1). The selected products come into contact with rain, seepage, or groundwater during their use phase, contain several compounds including organic substances, and are widely used in the

building and construction sector. The product groups tested comprised roofing membranes, varnishes, wood–plastic composites, sealants and coatings, grouts, cork granules, an infiltrative pavement, and foam glass aggregates. The three roofing membranes examined are used particularly to seal flat roofs and were made of polyisobutylene (PIB), ethylene–vinyl-acetate terpolymer (EVA), and ethylene–propylene–diene terpolymer (EPDM, production without mercaptobenzothiazole). Four varnishes were selected that contain different binders and that released substances with environmentally hazardous properties in previous tests [28]. The wood–plastic composites tested were terrace tiles of various composition. The sealants and coatings examined are commercially available as liquid products or pastes curing in situ and were selected based on different chemical bases. Areas of application are indoor and outdoor waterproofing

Table 1 List of products tested including product description and applied leaching test

Product group	Product code	Product description	Leaching test (eluate fraction tested)
Roofing membranes	DF4	Polyisobutylene (PIB)	DSLTL (1 + 2)
	DF6	Ethylene–vinyl-acetate–terpolymer (EVA)	DSLTL (1 + 2)
	DF7	EPDM (production without mercaptobenzothiazole)	DSLTL (1 + 2) and DSLTL (8)
Varnishes	F10	Swimming pool paint (chlorinated rubber)	DSLTL (1 + 2)
	F11	Colored lacquer for exterior (polyurethane dispersion)	DSLTL (1 + 2)
	F14	Clear lacquer and primer (acrylate dispersion)	DSLTL (1 + 2)
	F16	White lacquer (acrylate dispersion with polyurethane dispersion and alkyd resin emulsion)	DSLTL (1 + 2)
Wood–plastic composites	WPC1	60% wood, 35% HDPE, 5% additives	DSLTL (1 + 2)
	WPC2	75% wood fiber, polyethylene, additives, dyes	DSLTL (1 + 2)
	WPC3	60% natural fiber, 30% polyethylene, 10% additives	DSLTL (1 + 2)
Sealants and coatings	DM1	Latex sealing compound for shower sealing	DSLTL (1 + 2)
	DM2	Latex sealing compound for composite sealing under tile and panel coverings	DSLTL (1 + 2)
	DM3	2-component sealing, polyurethane-based	DSLTL (1 + 2)
	DM4	Sealant and adhesive based on hybrid polymer	DSLTL (1 + 2)
	DM5	Bitumen sealant for leaks in roofs	DSLTL (1 + 2)
	DM6	Liquid plastic for coating	DSLTL (1 + 2)
	DM7	Silicone sealant, neutral crosslinking, alkoxy system	DSLTL (1 + 2)
	DM8	Silicone sealant, acid crosslinking, acetoxo system	DSLTL (1 + 2)
	DM9	Acetate-based silicone sealant	DSLTL (1 + 2)
	DM10	Silane-modified polymer for repairing and sealing roofs and gutters	DSLTL (1 + 2)
Grouts	MOE1	2-component joint grout, epoxy resin-based	DSLTL (1 + 2) and percolation (L/S 2)
Cork granules	KG1	Without recycle content	percolation (L/S 2)
	KG2	Without recycle content	percolation (L/S 2)
	KG3	Without recycle content	percolation (L/S 2)
Water-bound pavement	WEG	Water-bound product, mix of an inorganic and an organic component	percolation (L/S 2)
Foam glass aggregates	SG1	Product with technical approval	percolation (L/S 2)
	SG2 + SG3	Different grain sizes mixed	percolation (L/S 2)

buildings and roof sealing. The grout examined is a two-component joint grout with epoxy resin as binder and is used to fill joints between tiles. The cork granules are used as filling material in the substructure of artificial turf pitches and are advertised as an ecological and renewable alternative to filling materials based on SBR, EPDM, or TPE [29]. A water-bound and water-permeable pavement created from two components, a mixture of a specific mineral aggregate and a stabilizer, was tested, as were two different mixtures of foam glass aggregates.

Leaching tests

In accordance with the requirements of the draft for CEN/TS 17459 [26], monolithic, sheet- or plate-like construction products were eluted using the dynamic surface leaching test (DSLTL, DIN CEN/TS 16637-2, status since 2021 prEN 16637-2 [19]) and granular products were eluted using the horizontal up-flow percolation test (CEN/TS 16637-3, status since 2021 prEN 16637-3 [20]). Depending on the type of product, both tests might be suitable for one product. For grout materials, for instance, two scenarios can be considered: water running off the surface and water percolating from the construction product. Therefore, the product MOE1 was leached using both the DSLTL and the percolation test.

Preparation of test specimens

The test specimens were prepared according to the manufacturers' instructions in the technical data sheets. The roofing membranes were cut to sizes of 26.3 cm 19 cm to fit best into the testing vessel (see 2.2.2). The varnishes were applied to the roughened surface of sandblasted glass plates (26.5 cm 19 cm). After drying under indoor air conditions, the test specimens were conditioned for 7 days at $21.5\text{ }^{\circ}\text{C} \pm 1.0\text{ }^{\circ}\text{C}$ and $60\% \pm 5\%$ relative humidity. The wood–plastic composites were cut in such a way that an exposed area of about 1000 cm^2 was obtained, taking into account the cavities accessible by water. Test specimens were prepared with 26.5 cm 7.1 cm 1.1 cm for WPC1, with 20 cm 14.5 cm 2 cm for WPC2, and with 15 cm 14.5 cm 2.2 cm for WPC3. The sealants and coating materials were applied to the roughened surface of sandblasted glass plates (26.5 cm 19 cm), first dried for 1–2 days under laboratory conditions and then conditioned at $21.5\text{ }^{\circ}\text{C} \pm 1.0\text{ }^{\circ}\text{C}$, $60\% \pm 5\%$ relative humidity for at least 4 weeks. The grout was prepared by pouring the mixture of the two components into a prepared mold (25 cm 14 cm 0.7 cm internal dimensions). The mold was lined with cling film beforehand to prevent the grout from sticking to its surface. The test specimen was kept under ambient air conditions and was removed from the mold after 4 days. Afterward, the test specimen was conditioned for 8 days at $21.5\text{ }^{\circ}\text{C} \pm 1.0\text{ }^{\circ}\text{C}$, $60\% \pm 5\%$ relative

humidity. To prepare the test material for the percolation test, the prepared mixture of MOE1 was spread on an HDPE sheet and dried under ambient conditions for 46 days. The dried material was gently deagglomerated (particles $< 1\text{ cm}$) to enable filling into the glass column.

For the percolation tests, the subsample quantities of grains introduced into the columns were representative of the total samples. According to prEN 16637-3, the grain fractions that are $> 22.4\text{ mm}$ must be crushed in such a way that at least 45% of the grain size is $< 4\text{ mm}$. This condition was fulfilled for all products examined in the percolation test, except for the cork granules. A grain size reduction of the cork granules was not feasible and meaningful (condition of the intended use).

Dynamic surface leaching test

For the DSLTL [19], all-glass aquaria (28 cm 20 cm 13 cm, internal dimensions) were used. Glass plates coated with varnishes or sealants and coatings on one side were placed on the bottom of the glass basins with the coated side facing upward. The test specimens of the other products were placed on glass carriers with a small contact area to enable water contact from all sides. For each product, one specimen was used per test, except for WPC1, whose smaller dimensions demanded two test specimens to fulfill the standard procedure's requirement for the minimum surface area to be tested. Deionized water (Milli-Q[®], conductivity $< 5\text{ }\mu\text{S cm}^{-1}$) was added at a liquid-to-surface area ratio (L/A) of 25 L m^{-2} (2.5 mL cm^{-2}), and the entire water volume was renewed after 6 h. According to prEN 16637-2, the DSLTL comprises eight leaching periods within 64 days. In this study, as recommended in CEN/TR 17105, only eluates from the first two leaching periods of 6 h and an additional 18 h (in total 24 h) were collected, combined, and then subdivided into aliquots for chemical analysis and ecotoxicity testing. A complete DSLTL was carried out for the DF7 roofing membrane to perform ecotoxicological tests and chemical analyses also for the eluate from the last leaching phase (eluate 8 from the period of 36–64 days). The DSLTLs were performed at $20\text{ }^{\circ}\text{C}$ to $22\text{ }^{\circ}\text{C}$ in the dark. A blank control was run with deionized water (Milli-Q[®]) in an additional leaching vessel for each test series.

Up-flow percolation test

For the percolation test [20], borosilicate glass columns with an inner diameter of 5.9 cm, 6.5 cm or 10 cm and a filling height of 26 cm to 40 cm were used, depending on material availability, maximum grain size, and desired eluate volume. After packing the columns with a representative sample of the granular product, the samples were exposed to an upward flow of the eluent (deionized water (Milli-Q[®]), conductivity $< 5\text{ }\mu\text{S cm}^{-1}$). The

flow rate of the eluent was calculated in accordance with DIN prEN 16637-3, considering the specified linear flow velocity of 30 cm d^{-1} and the selected column diameter. The saturation rate (flow rate to saturate the column before the actual start of the experiment) was calculated in such a way that the saturation was completed within 4 h. After saturation, flow was interrupted for 12 h to enable a pre-equilibration of the test sample. The percolation tests were carried out up to the accumulated eluate fraction at a liquid-to-solid ratio (L/S) of 2 L kg^{-1} , which was collected for chemical and ecotoxicological analyses. A blank control was run with deionized water (Milli-Q®) and the same amount of inflow and outflow filter material (washed quartz sand) in an additional column for each test series.

All leaching tests were performed at BAM. Aliquots of the eluates were frozen at $\leq -18 \text{ }^\circ\text{C}$ in either 50 mL PP (polypropylene) containers (for the algae, daphnia, luminescent bacteria, umu, and Ames fluctuation tests), or 150 mL PET (polyethylene terephthalate) containers (for the fish egg test), or annealed glass bottles (for chemical analysis, i.e., GC-MS and LC-ESI-QTOF screening). The samples were shipped in frozen condition to Hydrotox GmbH for ecotoxicity testing and to TZW for chemical analysis and stored at $\leq -18 \text{ }^\circ\text{C}$ until the start of the respective investigations. Ecotoxicity tests were started within 2 months after sampling the eluates.

Ecotoxicity tests

The ecotoxicity testing of the eluates comprised algae, daphnia, fish egg, luminescent bacteria, umu, and Ames fluctuation tests following the CEN/TR 17105 [25] guideline for ecotoxicological testing of construction products. The tests were conducted in accordance with ISO standards, applying the lowest ineffective dilution (LID) concept, and have been described in more detail by [23, 24].

Briefly, the algae growth inhibition test was performed with the algae species *Raphidocelis subcapitata* in accordance with ISO 8692:2012 [30]. The inhibition of growth was determined after 72 h by measuring the chlorophyll fluorescence. The acute daphnia toxicity test was carried out in accordance with ISO 6341:2012 [31] with *Daphnia magna* using synthetic dilution water. The mobility of the daphnids was evaluated after 24 h and 48 h. The fish egg test was carried out with eggs from *Danio rerio* in accordance with ISO 15088:2007 [32]. The fertilized eggs were exposed in 24-well plates at $26 \text{ }^\circ\text{C} \pm 1 \text{ }^\circ\text{C}$ for 48 h. The luminescent bacteria test was conducted in accordance with ISO 11348-2:2007 [33] using liquid-dried luminescent marine bacteria of the species *Aliivibrio fischeri*. The decrease in luminescence was determined after an exposure time of 30 min.

As genotoxicity tests, the umu and Ames fluctuation tests were applied. The umu test was performed in accordance with ISO 13829:2000 [34] with the bacterial strain *Salmonella typhimurium* TA1535/pSK1002. The bacteria were exposed for 2 h to the eluates with and without metabolic activation, followed by a growth phase of 2 h, after which the induction of the umuC gene was determined. The Ames fluctuation test was carried out in accordance with ISO 11350:2012 [35] with the bacterial strains *Salmonella typhimurium* TA 98 and TA 100 using the test kit from Xenometrics (Allschwil, Switzerland) in microtiter plates. The eluates were examined with and without metabolic activation after an incubation period of 48 h. The test is described in more detail in [36].

The LID corresponds to the lowest dilution factor D, at which effects below the specific threshold values were determined. The following effect-threshold values were applied for the LID: algae test 5%; daphnia test 10%; fish egg test 10%; luminescent bacteria test 20%; umu test: $\text{IR} < 1.5$; Ames fluctuation test: $\text{IR} < 2.0$ ($\text{IR} = \text{induction rate}$). In the four ecotoxicity tests, the eluates were tested from the dilution D2 (1:1 dilution of the eluate with test medium) on (dilution series: D2, D3, D4, D6, D8, D12, D16, etc.). It was assumed that the test organisms are not able to survive in the undiluted eluate, since the deionized water that is used as a leachant does not contain all required nutrients. For the genotoxicity tests, the lowest dilution tested was D1.5 in the umu test and D1 in the Ames fluctuation test. The blank control eluates were examined in the ecotoxicity tests with dilution D2, in the umu test with D1.5, and in the Ames fluctuation with D1. The blank control sample is intended to reliably exclude or detect possible artifacts due to contamination of the dilution water, the equipment, or the sampling vessels.

Chemical analysis

Basic analysis

The basic chemical analysis of the eluates included measurements of total organic carbon (TOC), total nitrogen (TN), pH, conductivity, and turbidity and was conducted immediately after sampling at BAM. For a detailed description of the applied methods, see Additional file 1 [37–40].

Inorganic anions and cations

The concentrations of anions in the eluates were determined by ion chromatography in accordance with DIN EN ISO 10304-1 [41]. For the analysis of cations, subsamples of the eluates were acidified with concentrated sulfuric acid to $\text{pH} < 2$ immediately after sampling. Concentrations of cations were determined by inductively coupled plasma optical emission spectrometry (ICP-OES, iCAP 7000 ICP-OES, Thermo Scientific) in

accordance with DIN EN ISO 11885 [42]. Cations with LOQs above LAWA GfS values (effect levels defined by the BUND/Länder-Arbeitsgemeinschaft Wasser) [43] and samples with very low concentrations were determined by inductively coupled plasma mass spectrometry (ICP-MS, iCAP Q equipped with a fast valve, Thermo Scientific) in accordance with DIN EN ISO 17294-2 [44]. Detailed information on the applied methods and limits of quantification (LOQ) are given in SI 2 and [36].

Screening of organic compounds

A combination of the three screening techniques GC-MS, LC-ESI-QTOF, and LC-DAD was applied. Analyses were performed at TZW for the following eluates: DF4, DF6, DF7 (both eluate 1+2 and eluate 8), F14, WPC1, WPC2, MOE1 (DSLIT and percolation test eluate), KG1, KG2, KG3, SG1, and SG2+SG3 (see Table 1). The screening for organic substances by GC-MS was performed in accordance with DIN EN 15768:2015-05 [45] after liquid/liquid extraction. An a priori approach (non-target analysis) was followed by evaluating and comparing the detected signals with the NIST database. Semi-quantitative analysis was performed using labeled internal standards. For LC-ESI-QTOF analysis, both a non-target analysis and a suspected-target analysis were carried out. In both approaches, the measured MS/MS spectra were compared with a spectra database for LC-QTOF systems for improved identification. In the case of LC screenings, no semi-quantitative assessments can be made. In addition, a liquid chromatographic method coupled to a UV detector (LC-DAD) was carried out. This method allows qualitative analysis by comparing the UV chromatograms of different samples. Details on the three analytical methods are given in SI 3 and in [36].

Quantitative analysis of selected organic substances

For the MOE1 eluate, the organic substances benzyl alcohol, 4-tert-butylphenol, 1-3-benzenedimethanamine, and *N,N*-dimethyl-1-dodecanamine were quantified by GC-MS after liquid-liquid extraction as described in [36]. These selected substances have been qualitatively detected and are partly labeled with H-phrases for environmental hazards. In eluates from the cork granules, selected phenols that were identified in screening tests and available as reference substances were quantified by BAM. Furthermore, isothiazolinones were quantified in the eluates from sealants and coatings. Details on the methods are given in SI 4.

Results

Ecotoxicity of eluates

The 27 construction products examined differed considerably in their ecotoxicological potential. Eluates

from nine products showed no effects in the four ecotoxicity and two genotoxicity tests. The eluates from the other 18 products caused ecotoxic effects ranging from low to very high effects (the higher the LID, the more toxic is the eluate) (Table 2). Very high ecotoxicities were particularly observed for the product groups of grouts and cork granules (up to LID = 16384 for the grout and LID = 24578 for the cork granules). Algae and luminescent bacteria tests were significantly more sensitive than the daphnia and fish egg tests in all tested eluates.

The eluates from the product groups of roofing membranes, varnishes, wood-plastic composites, and foam glass aggregates did not show ecotoxic effects in any biotest, or only low toxicity in the algae test (e.g., the varnish F14 with LID_A = 12). For the EPDM roofing membrane DF7, the eluate fraction 8 was tested in addition to the eluate fraction 1+2 to obtain a complete data set compared with the EPDM roofing membrane DF5, which was tested in a previous study [7]. While the DF5 roofing membrane showed significant effects in both fractions (LID 12 to 128), the product DF7 tested in this study showed no effects in the eluate fraction 1+2 and only a very low effect in eluate fraction 8 with LID_A = 3 in the algae test. The DF5 roofing membrane contains mercaptobenzothiazole, while the DF7 EPDM roofing membrane is produced without mercaptobenzothiazole.

For sealants and coatings, the two products based on latex (DM1, DM2) and the products DM8 and DM10 were particularly conspicuous in the algae test (up to LID_A = 128) and partly also in the luminescent bacteria test (up to LID_{lb} = 12). Preservatives such as isothiazolinones are suspected of being the cause. The sealing compounds based on PU, bitumen, or liquid plastic, on the other hand, were not very ecotoxic (LID ≤ 4).

The grout MOE1, eluted in both the DSLIT and the percolation test, caused particularly high ecotoxicities in all biotests. LID values of up to 3072 were observed in the DSLIT eluate and up to LID 16384 in the percolation test eluate. The sensitivity of the organisms was highest for luminescent bacteria, followed by the algae, daphnia, and fish eggs in both the DSLIT and percolation test eluate. The eluates were also mutagenic in the umu test. In all biotests, greater effects were observed in the percolation test eluate than in the DSLIT eluate.

The extremely high toxicities of the three cork granules eluates in the algae test were also particularly striking (LID_A values of 12288 to 24576). All three eluates were also ecotoxic for daphnia and luminescent bacteria and were mutagenic in the umu test (KG1) and in the Ames fluctuation test (KG2).

Table 2 Ecotoxicity of eluates

Product group	Product code	Leaching test (eluate fraction)	Algae LID _A	Daphnia LID _D	Fish egg LID _{Egg}	Luminescent bacteria LID _{lb}	Umu LID _{EU}	Ames
Roofing membranes	DF7	DSLTL (8)	3	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Varnishes	F14	DSLTL (1 + 2)	12	≤ 2	≤ 2*	≤ 2	≤ 1.5	1
Wood–plastic composites	WPC1	DSLTL (1 + 2)	6	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Wood–plastic composites	WPC2	DSLTL (1 + 2)	6	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Sealants and coatings	DM1	DSLTL (1 + 2)	64	≤ 2	≤ 2	12	≤ 1.5	1
Sealants and coatings	DM2	DSLTL (1 + 2)	48	≤ 2	≤ 2	3	≤ 1.5	1
Sealants and coatings	DM4	DSLTL (1 + 2)	4	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Sealants and coatings	DM5	DSLTL (1 + 2)	≤ 2–6	≤ 2	≤ 2	4	≤ 1.5	1
Sealants and coatings	DM8	DSLTL (1 + 2)	12	3	3	12	≤ 1.5	1
Sealants and coatings	DM9	DSLTL (1 + 2)	≤ 2	6	≤ 2	≤ 2	≤ 1.5	1
Sealants and coatings	DM10	DSLTL (1 + 2)	128	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Grouts ^a	MOE1	DSLTL (1 + 2)	384 1536	64	8	2048	3	1
Grouts ^a	MOE1	Perc (L/S 2)	3072	256 512	24 32	12288 16384	6	1
Cork granules	KG1	Perc (L/S 2)	12,288	24	≤ 2	24	6	1
Cork granules	KG2	Perc (L/S 2)	24,576	16	3	24	≤ 1.5	2
Cork granules	KG3	Perc (L/S 2)	12,288	24	≤ 2	24	≤ 1.5	1
Water-bound pavement	WEG	Perc (L/S 2)	24	≤ 2	≤ 2	16	3	16
Foam glass aggregates	SG1	Perc (L/S 2)	4	≤ 2	≤ 2	≤ 2	≤ 1.5	1
Foam glass aggregates	SG2 + SG3	Perc (L/S 2)	3	≤ 2	3	≤ 2	≤ 1.5	1
Blank tests			≤ 2	≤ 2	≤ 2	≤ 2	≤ 1.5	1

Results are given as lowest ineffective dilution (LID). Results are listed only for products with ecotoxic effects (LID higher than blank values)

^a Eluate was tested twice. Two values are listed when the tests resulted in different LID

The water-bound pavement (WEG) was especially conspicuous in the two mutagenic tests with LID = 3 in the umu test and with LID = 16 in the Ames fluctuation test.

Chemical characterization of eluates

Basic parameters

The basic parameters TOC, TN, pH, conductivity, and turbidity (Table 3) are useful indicators of the sum of leached substances in eluates and can give first hints about the possible ecotoxic potential of the eluates. The TOC is a sum parameter for the amount of leached organic substances in the eluates. High TOC values were observed in eluates from the grout MOE1, the three cork granules, and the water-bound pavement. All these products exhibit high ecotoxic effects. In contrast, eluates from roofing membranes, varnishes, wood–plastic composites, and foam glass aggregates contained low amounts of organic compounds, with TOC values of < 20 mg L⁻¹, and showed only low ecotoxic effects. The TOC values for the sealants and coatings varied with the product, ranging from very low TOC values of 0.8 mg L⁻¹ for DM8 and DM9 to TOC values of about 40 mg L⁻¹ for the two latex sealants DM1 and DM2. The ecotoxic effects tend to be higher for products from which

greater amounts of organic compounds were leached (e.g., DM1 and DM2).

The content of organic substances that contain nitrogen, as indicated by TN values in the eluates, was low in the products tested. The eluate from the grout MOE1 was an exception, with a TN of up to 270 mg L⁻¹ in the percolation test eluate.

The pH was between 6.3 and 8.4 for most products. Higher pH values (> 9) were observed in the eluates from the grout and the foam glass aggregates, while acidic pH values of 4.6 to 4.9 were measured in the eluates from the cork granules. The measured conductivity values varied widely between the products, indicating different amounts of leachable ions in the eluates. Especially high conductivity values and thus ion contents were observed in the eluates from the grout, the three cork granules, and the water-bound pavement. Again, all these products showed high ecotoxic potential. Turbidity was measured only in the percolation test eluates. While the grout and the foam glass aggregate eluates showed low turbidity (FNU < 10), the cork granule and pavement eluates showed greater turbidity, with values up to FNU = 108 for the eluate KG2, indicating greater amounts of mobilized colloids. In accordance

Table 3 Results of basic chemical analysis and analyzed anions of products with ecotoxic effects

Product group	Product code	Leaching test (eluate fraction)	TOC in mg L ⁻¹	TN in mg L ⁻¹	pH	Conductivity in μcm^{-1}	Turbidity FNU	F ⁻ in mg L ⁻¹	Cl ⁻ in mg L ⁻¹	Br ⁻ in mg L ⁻¹	NO ₂ ⁻ in mg L ⁻¹	NO ₃ ⁻ in mg L ⁻¹	PO ₄ ³⁻ in mg L ⁻¹	SO ₄ ²⁻ in mg L ⁻¹
Roofing membranes	DF7	DSLIT (8)	0.5	n.d.	8.4	2.5	n.d.				0.15	0.39	0.47	0.27
Varnishes	F14	DSLIT (1+2)	12.6	n.d.	7.5	9.3	n.d.				0.16		0.48	0.84
Wood-plastic composites	WPC1	DSLIT (1+2)	19.4	n.d.	6.3	39.3	n.d.		4.60	0.25	0.29	0.77		1.32
Wood-plastic composites	WPC2	DSLIT (1+2)	4.6	n.d.	6.8	8.2	n.d.					0.34	0.53	0.41
Sealants and coatings ^a	DM1	DSLIT (1+2)	38.4	3.3	8.4	137	n.d.		0.73		0.13		1.20	24.3
Sealants and coatings ^a	DM2	DSLIT (1+2)	41.7	5.1	8.0	165	n.d.		0.86	11.3	0.15	0.36	1.67	29.4
Sealants and coatings ^a	DM4	DSLIT (1+2)	13.8	0.8	7.8	5.9	n.d.						1.54	0.43
Sealants and coatings ^a	DM5	DSLIT (1+2)	28.9	2.3	7.3	90.6	n.d.		7.64		0.12		1.18	1.27
Sealants and coatings ^a	DM8	DSLIT (1+2)	0.8	<0.05	6.6	1.7	n.d.						1.04	0.49
Sealants and coatings ^a	DM9	DSLIT (1+2)	0.8	<0.05	7.3	1.7	n.d.							0.54
Sealants and coatings ^a	DM10	DSLIT (1+2)	7.6	0.4	6.4	10.0	n.d.						1.16	0.75
Grouts	MOE1	DSLIT (1+2)	427	81	9.1	240	n.d.	0.87	2.99	0.61	0.45	0.39	0.48	1.76
Grouts	MOE1	Perc (L/S 2)	1280	270	9.0	600	7.0	9.83	9.83	1.38	0.70	0.47	0.60	5.38
Cork granules	KG1	Perc (L/S 2)	2180	n.d.	4.6	599	23.0	2.08	22.5	1.93		0.52	34.0	2.88
Cork granules	KG2	Perc (L/S 2)	1420	10.3	4.9	591	108	1.30	25.6	2.16	0.32	0.55	41.4	1.04
Cork granules	KG3	Perc (L/S 2)	848	n.d.	4.8	433	20.5		15.0	1.22		0.34	50.0	1.73

Table 3 (continued)

Product group	Product code	Leaching test (eluate fraction)	TOC in mg L ⁻¹	TN in mg L ⁻¹	pH	Conductivity in μ cm ⁻¹	Turbidity FNU	F ⁻ in mg L ⁻¹	Cl ⁻ in mg L ⁻¹	Br ⁻ in mg L ⁻¹	NO ₂ ⁻ in mg L ⁻¹	NO ₃ ⁻ in mg L ⁻¹	PO ₄ ³⁻ in mg L ⁻¹	SO ₄ ²⁻ in mg L ⁻¹
Water-bound pavement	WEG	Perc (L/S 2)	273	5.9	6.9	863	16.6		7.89	0.18	0.17		1.50	33.3
Foam glass aggregates	SG1	Perc (L/S 2)	6.2	0.3	9.3	120	0.9	1.09		0.33	0.50	0.75	0.75	2.97
Foam glass aggregates	SG2 + SG3	Perc (L/S 2)	5.1	0.3	9.5	380	0.8	0.99	1.08	0.43	0.52	0.58	0.57	6.44

TOC total organic carbon, TN total nitrogen; concentrations of anions in the range of the LOQs (limits of quantification) are not included

^a Data from sealants and coatings are mean values from two leaching tests

with prEN 16637-3:2021, centrifugation of the eluates is required for samples of turbidity above 100 FNU.

For the grout MOE1, all basic parameters were higher in the percolation test eluate fraction L/S 2 than in the DSLT eluate fraction 1 + 2, indicating that percolation of granular material leads to greater exposure and thus greater leaching of substances.

Inorganic anions and cations

The inorganic components of eluates can be determined with comparatively simple analysis methods, and their possible environmentally hazardous effects can be derived using threshold values. The LAWA GfS threshold values for groundwater are used here as reference, although they do not apply directly to eluates [43]. The concentration of anions was low in most eluates (Table 3). Fluoride could be detected only in concentrations above the limit of quantification (LOQ) in the eluates from the grout, two cork granules, and the foam glass aggregates. Greater chloride concentrations were measured in the eluate from the sealant DM5, as well as in the percolation test eluates from the grout, the cork granules, and the water-bound pavement. The maximal value of 25.6 mg L^{-1} observed in the eluate KG2, however, is far below the threshold value of 250 mg L^{-1} for chloride [43]. The measured concentrations of bromide, nitrite, and nitrate were low in all eluates, except for a higher bromide⁻ concentration of 11 mg L^{-1} in the DM2 eluate. Significant amounts of phosphate were detected only in the three cork granules eluates with values of up to 50 mg L^{-1} . Sulfate concentration was elevated in eluates from the two latex sealants DM1 and DM2 and in the eluate from the pavement. Here, too, the maximal measured concentration of 33 mg L^{-1} was far below the threshold value of 250 mg L^{-1} for sulfate [43]. Overall, the measured concentrations of leached anions do not indicate possible ecotoxic effects.

In contrast, the concentrations of selected cations in several elements were above the LAWA GfS threshold values in eluates from specific products (Table 4, values above threshold values are marked in bold letters). In the eluates from the roofing membranes, the varnishes, and the wood–plastic composites, cation concentrations were mostly below LOQ or in a very low concentration range, except for a high copper value of $100 \text{ } \mu\text{g L}^{-1}$ in the WPC1 eluate and moderate sulfur concentration of $12.8 \text{ } \mu\text{g L}^{-1}$ to $40.6 \text{ } \mu\text{g L}^{-1}$. The cation concentrations for sealants and coatings were also inconspicuous, as they were low or only marginally elevated. An exception is the product DM2, for which the concentrations of As, Cu, Pb, and Zn considerably exceeded the LAWA GfS threshold values. In the grout MOE1, the percolation test eluate showed higher values for Cu, Ni, Pb, and Zn. Like for the basic

parameters, the DSLT eluate contained lower concentrations of all elements than the percolation test eluate. In general, more elements with higher concentrations were measured in the percolation test eluates in all products. Especially high concentrations of several elements were determined for the cork granules. Here, boron, barium, copper, nickel, lead, and zinc exceeded the LAWA GfS threshold values. The water-bound pavement and the foam glass aggregates eluates showed suspicious concentrations of arsenic, cadmium, and vanadium. In the pavement, the nickel and zinc values were additionally elevated at $92 \text{ } \mu\text{g L}^{-1}$ and $138 \text{ } \mu\text{g L}^{-1}$, respectively. Although, for instance, the foam glass aggregate eluates contained significant amounts of As, B, Cd, Pb, and V exceeding the threshold values used as reference, the eluates showed only a very low ecotoxicity of $\text{LID} < 4$.

Identification of organic compounds

To identify organic substances, both target and non-target analyses were carried out. While extensive libraries on identifying organic compounds are available for GC–MS screenings, the number of LC spectra is lower. The GC–MS screening was followed using an a priori approach (non-target analysis): the highest signals in the chromatogram were selected and then matched with the GC_{NIST} database. For the LC–MS analysis, both suspected-target analysis and an a priori approach were performed. For the suspected-target analysis, a comparison with two substance lists (roofing membranes and varnishes) was first carried out with the data sets obtained. However, these substance lists were derived from GC analysis and are, therefore, only partially compatible with the LC method. Therefore, an a priori approach for peak extraction in combination with the LC_{NIST} database was used as a further strategy for identifying the samples.

Using the same methods and instruments and evaluating the results by the same employees, there should be sufficient comparability between the samples to derive trends, even though the types of samples vary greatly from foam glass aggregates to cork granules. Considerable differences can be observed in the sum of organic substances detected in each sample examined (Fig. 1a). The number of substances found ranged from 10 in the roofing membrane DF7 to almost 60 in the cork granules KG1. No general trend that more signals were found in eluates with high ecotoxicity was observed. For the wood–plastic composites, for instance, the number of detected organic substances is high, although their ecotoxicological potential is rather low. The number of hits within the group of cork granules also differed considerably, while their ecotoxic effects are in a similar range.

Plotting the same data with a normalized y-axis clearly shows that the number of detected substances was

Table 4 Selected cations of eluates from products with ecotoxic effects

Product group	Product code	Leaching test (eluate fraction)	As in $\mu\text{g L}^{-1}$	B in $\mu\text{g L}^{-1}$	Ba in $\mu\text{g L}^{-1}$	Cd in $\mu\text{g L}^{-1}$	Cu in $\mu\text{g L}^{-1}$	Mo in $\mu\text{g L}^{-1}$	Ni in $\mu\text{g L}^{-1}$	Pb in $\mu\text{g L}^{-1}$	V in $\mu\text{g L}^{-1}$	Zn in $\mu\text{g L}^{-1}$
LAWA GfS			3.2	180	175	0.3	5.4	35	7.0	1.2	4.0	60
Roofing membranes	DF7	DSLIT (8)			4.06							23.4
Varnishes	F14	DSLIT (1 + 2)					1.98					21.7
Wood-plastic composites	WPC1	DSLIT (1 + 2)			4.43		110					40.6
Wood-plastic composites	WPC2	DSLIT (1 + 2)			1.69		6.74					12.8
Sealants and coatings ^a	DM1	DSLIT (1 + 2)	1.06		3.14	0.11	4.17	1.04	10.3	1.17		
Sealants and coatings ^a	DM2	DSLIT (1 + 2)	2.04		10.75	0.16	110	0.53	3.26	47.5		84.2
Sealants and coatings ^a	DM4	DSLIT (1 + 2)			0.82	0.07	5.74			0.37		
Sealants and coatings ^a	DM5	DSLIT (1 + 2)	0.19		2.69	0.15	2.54	0.59	4.84	1.05		34.7
Sealants and coatings ^a	DM8	DSLIT (1 + 2)		17.2	1.99		4.87		18.5			
Sealants and coatings ^a	DM9	DSLIT (1 + 2)		15.3	1.38		6.88					
Sealants and coatings ^a	DM10	DSLIT (1 + 2)			2.2		3.53		10.4			
Grouts	MOE1	DSLIT (1 + 2)	0.15		2.01		7.89		5.42	3.89		105
Grouts	MOE1	Perc (L/S 2)	0.78		25.8		38.6	4.85	8.72	6.74	2.85	236
Cork granules	KG1	Perc (L/S 2)	1.32	583	999		60.4		9.24	1.7	1.81	71.1
Cork granules	KG2	Perc (L/S 2)	0.82	706	1370		33.9		8.52	3.78	3.75	38.6
Cork granules	KG3	Perc (L/S 2)	0.68	612	1380		58.0		10.2	3.42	3.05	77.5
Water-bound pavement	WEG	Perc (L/S 2)	408		130	6.1	27.1	5.2	9.2	0.8	14.9	138
Foam glass aggregates	SG1	Perc (L/S 2)	186	732		2.54	4.16	3.3	10.8	1.28		
Foam glass aggregates	SG2 + SG3	Perc (L/S 2)	178	595		3.26	4.46	13.6	5.66	1.73	4.28	

Selection criteria: elements that are included in the LAWA list of threshold values (LAWA GfS, "Geringfügigkeitsschwellenwert"; English "insignificance threshold") from 2016 [42]; concentrations in the range of the LOQs (limits of quantification) are not included

^a Data from sealants and coatings are mean values of two leaching tests

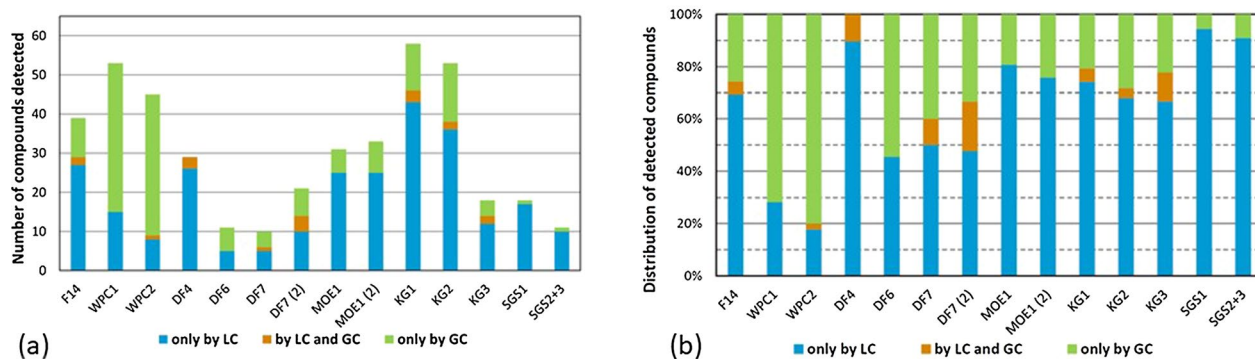


Fig. 1 Summarized results from screening organic compounds by GC–MS- and LC–MS-techniques. **a** Absolute hits of organic compounds in all samples examined; **b** Relative distribution of hits of the two MS methods in all samples examined. DF7 (2) = eluate fraction 8, MOE1 (2) = eluate from percolation test

generally higher for LC–MS than for GC–MS, except for the wood–plastic composites and the DF6 eluates (Fig. 1b). Substances found with both GC–MS and LC–MS are depicted in orange. Their share of the total number of hits is around 5%, and is thus rather small. This result underlines that the two techniques (LC and GC) and approaches (non-target and suspected-target approaches) can provide additional information. Detailed information on the screening results, including completed lists of substances detected, is given in [SI 5, 24].

In addition to the MS techniques, the samples were also analyzed by reversed phase separation coupled with photometric detection (LC–DAD). The chromatograms obtained were evaluated only qualitatively and no attempt was made to elucidate signals with reference standards. Nevertheless, valuable information can be obtained from these measurements. UV detects mainly unsaturated and aromatic compounds. Clear differences can be found between the samples. For example, the extracts from the cork granules contained a broad large signal in the middle retention range, while less was found in the foam glass aggregates. Differences between the DSLT and percolation test eluates from the MOE1 grout can also be observed and coincide with the trends in the screening and ecotoxicity results. Examples of this method can be found in SI 5.

Linking ecotoxic effects and the chemical composition of eluates

To link the results of the ecotoxicological investigations with those of the chemical analysis, the GHS classifications of the identified substances with regard to environmental hazards (acute: H400, chronic H410, H411, H412) were searched in the ECHA database [46]. In a second step, the ecotoxicity data for fish, daphnia, and algae for

all substances classified in H400, H410, H411, and H412 were extracted from the ECHA database [46]. Toxicity data on luminescent bacteria are not systematically recorded under REACH and was, therefore, not included in the research.

In all the samples examined, at least one compound classified with 400 s H-statement was found (data presented in SI 6). Some of the substances identified have very high ecotoxicities in the $\mu\text{g L}^{-1}$ range. However, determining the cause of ecotoxicity from these data is possible only to a very limited extent for the following reasons:

- The qualitative analysis of the identified substances does not allow any statement on their concentration.
- Although great effort was made by applying two mass-spectrometry-based screening methods targeting different substances, the samples might contain other compounds that were not detected, due to the selectivity of the enrichment and the analytical methods.
- Not all signals could be identified, as references are missing in the available databases.
- A large number of the substances identified are not covered by REACH, and therefore, classifications and ecotoxicity data are not available.

Despite these constraints, the organic screening data give first hints about the presence of potentially toxic substances and can be used to prioritize substances for which a quantitative analysis seems to be meaningful. Hence, to reasonably link ecotoxic effects with specific substances, both their ecotoxic potential and their concentration must be known.

Within the scope of this study, only a few suspicious substances that were identified with the screening

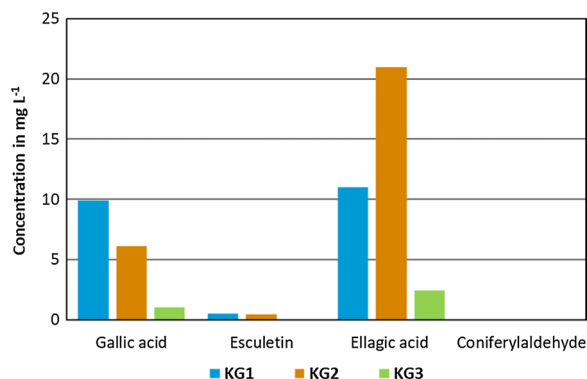


Fig. 2 Concentration of selected phenols in cork granules (KG 1 to 3)

methods could be quantitatively analyzed. Among these were phenols in the very ecotoxic eluates from the cork granules (Fig. 2). Of the four analyzed phenols, the highest concentration of up to 21 mg L⁻¹ was observed for ellagic acid, followed by gallic acid and very low amounts of esculetin. Coniferylaldehyde was not detected in any of the three eluates. For KG1, both gallic and ellagic acid were measured at around 10 mg L⁻¹, while for KG2 the content of ellagic acid was threefold higher than for gallic acids. In KG3, the lowest amounts of all three phenols were determined. Differences in the concentrations of these investigated phenols, however, did not cause any significant differences in the observed toxicities. In accordance with the ECHA registration dossier, gallic acid has a high algae toxicity (EC50 (72 h) = 2.19 mg L⁻¹, EC10 (72 h) 1 mg L⁻¹ to 1.5 mg L⁻¹, *Selenastrum capricornutum*). Although the concentrations of around 10 mg L⁻¹ measured in the undiluted eluates are 10 times higher than the EC10, they cannot come close to explaining the observed very high algae toxicity. No registration

dossiers are available for the other phenols. However, it is generally known that phenols excreted by plants, such as ellagic acid, gallic acid, and tannic acid, have significant algae toxicities [47]. The mixture of different phenols can, therefore, at least partially be suspected to cause the extremely high algae toxicity in the eluates from the cork granules.

In the eluates from sealants and coatings, isothiazolinones were analyzed, as they were suspected to cause the samples' toxicity. The maximum concentrations were 0.43 mg L⁻¹ for MIT (in DM1 eluate), 1.82 mg L⁻¹ for BIT (in DM1 eluate), and 0.45 mg L⁻¹ for OIT (in DM8 eluate). The concentrations of CMIT and DCOIT were below the detection limits of <0.02 mg L⁻¹ in all eluates, although for some products, these compounds are declared as ingredients. It is possible that these compounds are leached in such small quantities that they could not be detected with the analytical method applied. In accordance with the data on isothiazolinones in the ECHA registration dossier, algae are the most sensitive group of organisms (NOEC approx. 0.05 mg L⁻¹). This means that an ecotoxicity of up to approx. LID_A = 32 can be explained by the concentration of the preservatives.

To use chemical analysis to robustly estimate the overall ecotoxicological potential of a sample, ideally the ecotoxic properties and concentrations of all toxic substances present in the samples need to be known. Based on these data, the ecotoxicity can be estimated, applying the concept of mixture toxicity [48]. For the DSLT eluate from the grout MOE1, an attempt to identify and quantify the most relevant substances causing the ecotoxic effects on algae was implemented using concentration addition (Table 5). For this purpose, the concentrations of four selected substances were measured, and their NOEC

Table 5 Quantitative analysis of selected substances from DSLT eluates from the grout MOE1 and calculation of mixture toxicity

Substance	Measured concentration in mg L ⁻¹	NOEC algae in mg L ⁻¹	Calculated LID (concentration/NOEC)	EC50 algae in mg L ⁻¹	Pi	Pi/EC50
N,N-Dimethyl-n-dodecylamine	0.4	0.0026	153.8	0.016	0.006	0.368
4-tert-butylphenol	7.6	0.32	23.8	13	0.112	0.009
Benzyl alcohol	6.4	310	0.02	770	0.094	0.0001
1,3-Benzenedimethanamine	42.9	6.3	6.8	12	0.632	0.053
Sum	57.3	–	184.4	–	–	0.43
Measured LID in this study:			384–1536			
Mean measured LID from round robin test			166			2.33 Vol.-%
Calculated EC50 mixture toxicity						1.16 Vol.-%
Geometric mean of measured EC50 from round robin test						

Selection criteria for substances: confirmed by reference substances and/or substances classified as H400, H410, or H411. Measured concentrations are mean values of the eluate from this study and from the eluate for the round robin test [23]. NOEC and EC50 for algae are from the ECHA database [46]. Pi is the relative proportion of the i-th component in the mixture. The EC50 mixture toxicity is calculated as the reciprocal sum of Pi/EC50 values (1/0.43 = 2.33 Vol.-%)

and EC50 were derived from the ECHA database [46]. The mixture toxicity for the LID approach was calculated by dividing the measured concentration of the substances by their respective NOEC value reported in the ECHA database and summing these up. A ratio of above “1” indicates that the measured concentration is higher than the NOEC and that the calculated LID mixture toxicity corresponds to the dilution factor required to fall below significant ecotoxic effects. The EC50 mixture toxicity was calculated as the reciprocal sum of $P_i/EC50$ values. It should be noted that the use of NOEC data (and thus LID) is not highly recommended for the concept of mixture toxicity, since mixture toxicity is better estimated on the basis of clear effects, in particular via EC50 values. Nevertheless, the calculated LID mixture toxicity of 185 is in the same range as the measured LID of 384 to 1536 (dilutions of D384, D512, D768, D1024, and D1536 were tested and all resulted in similar inhibition of algae growth) and thus gives a plausible result. Given the mean LID of 166 determined for the same product in the round robin test [23], the calculated mixture toxicity of the four quantified substances comes very close to the mean measured LID. The EC50 mixture toxicity of the four substances of 2.33 vol.-% is also very similar to the geometric mean of the measured EC50 value from the round robin test of 1.16 vol.-%. The approach of mixture toxicity seems thus to reasonably explain the algae toxicity of the MOE1 eluate.

Discussion

Ecotoxic potential of construction products and its comparability

The construction products examined in this study covered a broad range of different product types and product groups that are typically used in the building sector and come into contact with water. For 2/3 of the products tested, ecotoxic effects were observed in at least one of the ecotoxicity tests. In addition, in previous studies that investigated eluates from other construction products, many products were proven to have ecotoxic effects [4–14, 49, 50]. The risk of causing harmful environmental effects through the leaching of toxic substances, therefore, seems to be relevant for a large part of construction products.

The extent of the ecotoxicological effects, however, varied considerably, ranging from low ($LID < 4$) to very high ecotoxicity (LID up to 24578) for the products tested in this study. Several of the products showed effects only in the algae test, while other products caused effects in all ecotoxicity tests. For most products, the sensitivity of the test organisms was greatest for algae, followed by luminescent bacteria, daphnia, and fish eggs. Comparing this sensitivity pattern with other studies [22, 23, 36, 50, 51],

algae and luminescent bacteria tend to be more sensitive than daphnia and fish eggs to the eluates from construction products. However, it is not recommended to focus ecotoxicity testing only on these two tests, since the sensitivity pattern might be different for other construction products and ecotoxicity assessment usually uses a test battery of organisms representing different trophic levels.

Surprisingly, the highest LID values were observed in the product group of cork granules, which are advertised as natural, environmentally friendly filling material for artificial turf pitches and playgrounds. All three cork granules tested caused extremely high algae toxicity ($LID_A = 12288$ to 24578). High ecotoxicity can thus not only be caused by substances leaching from synthetic building products, but also from natural materials. Cork consists mainly of the biopolymer suberin, lignins, and polysaccharides, but also contains about 15% of extractable components, including numerous phenols [52]. Various phenols in cork are described as water-soluble substances that can have antibacterial and insecticidal effects and serve the plants as natural repellents [53]. The analytical screening for organic substances in the cork eluates resulted in many signals, only some of which could be identified. The measured concentration of selected identified phenols (up to 27.5 mg L^{-1} in total for KG2 eluate), however, can explain only a small fraction of the overall very high ecotoxicity. Since very high TOC values of up to 2180 mg L^{-1} were determined in the cork granule eluates, it is clear that only a small fraction of the leached organic substances have been chemically analyzed so far. To elucidate the cause of the extraordinary high algae toxicity of the cork granules, further studies are needed to analyze these complex eluates. For a comprehensive environmental assessment, it is also important to know the fate of the leached substances. One way to obtain information about the fate of organic matter is with biodegradation tests, as recommended by draft CEN/TS 17459, if the TOC of an eluate is $> 10 \text{ mg L}^{-1}$ [25, 26, 54]. These tests are based on well-established OECD-ready biodegradability test guidelines, but were not the subject of the present study.

Previous studies on other materials that are applied in artificial turf pitches and outdoor sports floors showed that these materials also have significant effects in various ecotoxicity tests [50]. The EPDM granules (BAM-G1) and TPES granules (BAM-G2) eluted with the one-stage batch test in accordance with ISO 12457-1 resulted in LID values of up to 1536 and 64, respectively. The DSLT eluate from a water-permeable sports floor (BAM-14) made from these granules caused LID values of up to 192. A comparison of the ecotoxicological potential of different product types that can be used for the same application is needed to be able to choose the product with

the lowest environmental impact. A combination of the standardized elution test and ecotoxicity tests is a useful tool for this comparative consideration and is already applied for certain product groups in the eco-labeling criteria of the Blue Angel [55–57] and in European Assessment Documents (EAD) [58, 59]. Materials from natural sources should be examined in accordance with the same standards as are applied to chemical or artificial products, since they also might cause ecotoxic effects, as shown for the cork granules.

Within the product group of roofing membranes, the ecotoxicological potential clearly varied with the product type. The three products tested in this study (materials: PIB, EVA, and EPDM) were all inconspicuous. This was especially surprising with the EPDM roofing membrane DF7, since previous studies observed significant ecotoxic effects for DSLT eluates from other EPDM roofing membranes [7, 50]. Chemical analyses of leached pollutants in eluates from sixteen roofing materials including EPDM, PVC, TPO, EVA, PU, and bitumen membranes also indicated that EPDM roofing materials have the highest potential to contaminate roof runoff, mainly due to high zinc leaching [60]. In contrast to the EPDM product tested in previous studies, the production of the EPDM roofing membrane DF7 is conducted without mercaptobenzothiazole. Using an alternative production process, it seems to be possible to design an EPDM roofing membrane that is not ecotoxic. Since the ecotoxicological potential varies considerably within the product group of roofing membranes, this group is well-suited for eco-labeling to award the products with the lowest environmental impact.

Another product group with a broader range of observed ecotoxicity includes the sealants and coatings, which are also well-suited for eco-labeling. A Blue Angel eco-label was recently established for roof and sealing sheets [56]. The varnishes seem to be a less critical product group, since most eluates examined did not show significant ecotoxicity in any test system. However, there are special epoxy resin-based and polyurethane-based [61–63] varnishes in fire protection coatings [51] and corrosion inhibitors, in whose eluates some significant ecotoxicities were found.

The very high ecotoxicity of eluates from an epoxy resin-based 2-component grout (MOE1) was particularly striking. This product was examined in the DSLT and percolation tests, since (a) runoff from the surface of the joints and (b) percolation of the water-permeable joints are possible scenarios in which substances can be leached from the material. This example highlights that for some construction products not only one of the two harmonized leaching tests might be appropriate depending on the characteristics of the material and on the intended

application. Investigations of the MOE1 eluates from the two different leaching methods revealed that, for all biotests and chemical parameters (TOC, TN, conductivity, anions, and cations), higher values were observed in the percolation test eluate than in the DSLT eluate. These data were confirmed by further leaching and ecotoxicity tests conducted with the product MOE1 in the international round robin test [23] and is described in more detail in [36]. Higher concentrations of leached substances and consequently also greater ecotoxicities in the percolation test eluate than in the DSLT eluate can likely be explained by differences in the contact area and contact time of material and water for the two leaching methods. The different results for ecotoxicity and chemical analysis in eluates from the same construction product make it clear that the type of leaching test affects the amount of leached substances and thus plays an important role in interpreting and comparing the results. Transferring ecotoxicity data from one leaching test eluate to another is, therefore, possible only to a limited extent, and the applied leaching test should be known to interpret the ecotoxicity data from eluates. The application scenario has to be taken into account when choosing the leaching test. Comparing the ecotoxicity data determined for DSLT eluates from various grout types shows that the grouts based on epoxy resin, such as MOE1, exhibit a higher ecotoxicological potential than grouts made of modified cement, polyurethane, or polybutadiene [36].

Chemical characterization of eluates

Chemical analyses of eluates comprised basic parameters (TOC, TN, pH, conductivity, and turbidity), inorganic anions and cations, and screening for organic substances with LC–DAD, GC–MS, and LC–MS. By applying several standardized methods targeting different substances, the eluates could be characterized in a way that allows comparison of the eluates and gives first hints on possible reasons for the ecotoxicity observed.

The eluates with high TOC, such as those from grout or cork granules, exhibited high ecotoxicity. Although being an unspecific sum parameter, high TOC values may indicate the leaching of potential hazardous organic compounds from construction products. The concentration of anions in the eluates was low in all eluates and did not indicate possible ecotoxic effects. In some eluates, the concentrations of selected cations were above LAWA GfS threshold values that were defined for groundwater. However, considering the safety factors that were applied to define these threshold values, the measured concentrations of cations are unlikely to cause significant ecotoxic effects. Exceptions might be the eluates from the cork granules, the sealant DM2, the water-bound

pavement, and the percolation test eluate from MOE1, in which the concentration of several cations exceeded the threshold values significantly and thus might have contributed to the overall ecotoxicity of the eluates. For instance, the lead concentration in the eluate from the sealant DM2, measured at $48 \mu\text{g L}^{-1}$, was in the range of the reported effect concentrations for the most sensitive species of algae (EC_{50} (72 h): 20.5 to $364 \mu\text{g L}^{-1}$) [46]. On the other hand, the eluates from the foam glass aggregates, for which the concentration of several cations was also considerably higher than the LAWA GfS threshold values, caused only very low ecotoxicity. Thus, elevated concentrations of possibly toxic compounds do not necessarily lead to a toxic effect of the eluate. Overall, for most products tested, the leached anions and cations are not the reason for the observed toxicity. The ecotoxicity of eluates must, therefore, have been caused mainly by the content of organic substances that were leached from the construction products.

To investigate the complex mixture of leachable organic substances, various analytical techniques and strategies can be applied. In this study, a combination of GC-MS and LC-QTOF and a target and non-target approach were applied to identify many of the leached organic compounds. The two methods differ in their selectivity; the GC-MS targets volatile compounds with a more lipophilic character, while LC-MS enables the detection of substances with a wide polarity range and a wide molecular mass range [64]. The small, approximately 5% overlap of substances that were identified through both techniques clearly shows that the combination of both methods significantly expands the measurable spectrum, and neither of the two methods alone would lead to comparable results. The different specificity of the individual methods thus makes it useful to combine different analytical methods. The identification of the components of complex mixtures from leaching experiments is currently a major challenge for analytical chemistry. For example, in samples from leaching tests with plastic products, up to several thousand signals were found, of which an average of about 8% could be provisionally assigned to a structure [65]. Similarly, for the eluates from the construction products, many signals were found that could not be assigned to a structure, as the usual spectral libraries contain mass spectra for only some of the substances leached from construction products. The implementation and standardization of non-target analysis methods, including the procedure for data filtering and interpretation, is still a pending task.

Nevertheless, the screening data obtained for the analyzed samples clearly proves that various organic substances can be found in the eluates from construction products. The standardization of the applied methods

(including sample preparation, measurements with the same instruments, and evaluation by the same persons) makes the different eluate samples comparable enough to derive trends from the screening data. The number of hits found for organic compounds was highest for the eluates from the cork granules KG1 and KG2. This corresponds with the high ecotoxicity presumably caused mainly by organic compounds. In the eluate from cork granules KG3, however, a significantly lower number of hits was found, although its ecotoxicity is in the same range as that of the eluates KG1 and KG2. For wood-plastic composites, a high number of hits was found, although both the TOC values ($<20 \text{ mg L}^{-1}$) and the toxic potential ($\text{LID}=6$) were comparably low. A stringent link between the number of hits and the total carbon content and the ecotoxicity of the eluate is hence not given. It is striking that most substances in the wood-plastic composites were detected by GC-MS, indicating a high number of volatile compounds. These compounds might have evaporated during the biotests, leading to shorter exposure times and thereby possibly to lower ecotoxic effects.

Linking ecotoxicity and the chemical analysis of eluates

Many construction products consist of a complex mixture of several substances that can be leached to varying degrees and that have different potentials to affect the environment. It is, therefore, very challenging to assign ecotoxic effects to specific substances. Vice versa, deducing the ecotoxicity of an eluate based on chemical analysis is also a difficult and highly labor-intensive task, since this requires extensive analysis of the leached substances and knowledge of their ecotoxicological potential and their concentrations. In any case, the ecotoxicological potential of individual substances needs to be known to link ecotoxicity and chemical data. This information, however, is available only for some of the substances that have been registered under REACH. In particular, the reported H-phrases can be used to establish a link between the chemical analysis and the ecotoxic effects observed in a sample. Substances for which entries with H-phrases exist can be prioritized for the quantification of compounds in such samples. For instance, among the identified substances for the grout MOE1, *N,N*-dimethyl-1-dodecylamine stands out due to its very high algae toxicity (EC_{10} approx. $3 \mu\text{g L}^{-1}$). Other substances identified in this eluate sample and prioritized for quantification based on their reported hazard potential were benzyl alcohol, 4-*tert*-butylphenol, and 1,3-benzendimethanamine. Quantification of these substances and calculation of the mixture toxicity showed that a large part of this eluate's algae toxicity can be explained by the concentrations of these four substances and that *N,N*-dimethyl-1-dodecylamine is by far the most

important component. Calculating the mixture toxicity is hence a promising approach to narrow down the cause of ecotoxicity in eluates and link the ecotoxicity tests with chemical analysis. Nevertheless, the results should be evaluated with caution, since the concentrations of numerous other substances are not known. As stated above, ideally the concentrations and ecotoxic properties of all toxic substances present in the samples need to be known to robustly assign the overall ecotoxic effects to specific substances. However, this is usually not feasible, since the applied analytical methods might not detect or identify some of the leached substances, and data on their ecotoxicological potential are missing. In the case of the cork granules, for instance, the phenols gallic acid and ellagic acid, which were found in high quantities and are reported to be ecotoxic, can explain only a very small fraction of the observed algae toxicity. These eluates contain a particularly complex mixture of components, many of which probably cause the very high ecotoxicity. The analytical challenges involved in the ecological risk assessment of mixtures of unknown and variable composition are currently often addressed using the whole mixture approach [66]. This means they are assessed as if they were a single chemical. The whole mixture approach provides adequate information for product use. However, for an ecological improvement of product recipes, a component-based approach is important.

Conclusion

Determining the ecotoxicity of eluates from construction products records the summary effect of all leachable substances and is seen as a useful instrument for detecting the release of hazardous substances and for identifying products compatible with the goals of the Zero Pollution Ambition. Standardizing leaching procedures and biotests is important for making the results comparable. The choice of leaching test depends on the product type and its application scenario and affects the amounts of leached substances. The ecotoxicity results should, therefore, always be interpreted in the context of the applied leaching test. The benefit of ecotoxicity tests extends in particular to complex organic products with largely unknown ingredients. The results of ecotoxicity testing allow a prioritization of the eluates that should be subjected to detailed chemical analysis. Analysis costs can thereby be saved. Assigning ecotoxic effects to certain substances in the complex mixture of leached components is challenging and requires a broad analytical approach as well as knowledge of the ecotoxicological potential of the identified substances. Organic screening with a combination of GC–MS and LC–MS and ranking the identified substances based

on recorded hazard classification is a suitable approach to identify the relevant toxic substances that should be subjected to further quantifying analysis. The biological and the chemical–analytical approach thus complement each other in an ideal way to characterize the hazard potential of the eluates from construction products and to identify the environmentally hazardous components in these eluates. Organic substances leached from building surfaces with rain events form mixtures in the environment. So far, little research has been done on mixture toxicity and the joint effects of water-soluble organic chemicals commonly used in construction products. Our results confirm that, to move toward a more sustainable way of building and to reduce toxic effects in the environment, the combination of harmonized leaching and ecotoxicity tests is a reasonable approach that should be applied to construction products potentially exposed to water.

Abbreviations

BAM	Bundesanstalt für Materialforschung und-prüfung
BIT	Benzisothiazolinone
CEN/TC	European Committee for Standardization/Technical Committee
CEN/TR	European Committee for Standardization/Technical Report
CEN/TS	European Committee for Standardization/Technical Specification
CMIT	Chlormethylisothiazolinone
CPR	Construction Products Regulation
DCOIT	Dichloroethylisothiazolinone
DE-UZ	Umweltzeichen Blauer Engel
DIN	Deutsches Institut für Normung
EN	European Standard
DSL	Dynamic surface leaching test
EAD	European Assessment Document
EC50	Volume percentage causing 50% effect
ECHA	European Chemicals Agency
EPDM	Ethylene propylene diene-monomer rubber
ESI-QTOF	Electrospray ionization–quadruple-time-of-flight mass spectrometer
EVA	Ethylene–vinyl-acetate terpolymer
FPO	Flexible polyolefin
GC	Gas chromatography
HDPE	High-density polyethylene
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectrometry
ISO	International Organization for Standardization
LAWA	Bund/Länder-Arbeitsgemeinschaft Wasser
LC	Liquid chromatography
LC–DAD	Liquid chromatography coupled with a diode array detector
LID	Lowest ineffective dilution
LOQ	Limit of quantification
L/A	Liquid-to-surface area ratio given in L m ⁻²
L/S	Liquid-to-solid ratio given in L kg ⁻¹
MIT	Methylisothiazolinone
MS	Mass spectrometry
n.d	Not determined
NOEC	No observed effect concentration
OIT	Octylisothiazolinone
PET	Polyethylene terephthalate
PIB	Polyisobutylene
PP	Polypropylene
PU	Polyurethane
PVC	Polyvinyl chloride
REACH	Registration, Evaluation, Authorization of Chemicals

SBR	Styrene–butadiene rubber
TOC	Total organic carbon
TN	Total nitrogen
TPE	Thermoplastic elastomers
TPO	Thermoplastic polyolefins
TZW	DVGW Technologiezentrum Wasser

Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s12302-023-00711-w>.

Additional file 1: The following supporting information can be downloaded at: <https://enveurope.springeropen.com/>, SI 1: basic analysis methods; SI 2: inorganic anions and cations methods; SI 3: organic screening methods; SI 4: Quantitative analysis of selected organic substances; SI 5: Results of organic screenings; SI 6: classification and ecotoxicity data of identified substances.

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Author contributions

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Declarations

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Not applicable.

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