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Recommendation for a test battery for the ecotoxicological evaluation of the environmental safety of construction products

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HIGHLIGHTS

- 20 construction products have been submitted to different leaching tests followed by ecotoxicity testing.
- Low to very high ecotoxicity was observed in the algae, daphnia, luminescent bacteria, and fish egg test.
- Biodegradability of most eluates with significant TOC was acceptable.

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ABSTRACT

The European Construction Products Regulation allows Member States to adopt rules for evaluating the environmental impact of their buildings. The aim of the project was to develop recommendations for a test battery for the ecotoxicological assessment of the environmental impact of construction products for outdoor use and contribute to the European harmonization of test methods. From a shortlist of 39 products 20 products were included in the ecotoxicological testing program. Monolithic and plate-like construction products were eluted in the Dynamic Surface Leaching test (DSLT) in accordance with CEN/TS 16637-2, granular products were eluted in a one stage batch test in accordance with DIN EN 12457-1. The eluates were examined in four aquatic toxicity tests (algae, daphnia, luminescent bacteria, fish eggs), a genotoxicity test (umu test) and in the respirometer test (OECD 301 F). Here, low to very high ecotoxicity was observed (up to a dilution factor of 1536). Six out of 8 eluates, whose TOC exceeded 10 mg L^{-1} showed a good biodegradability above 75%. The intra-laboratory repeatability of the Lowest Ineffective Dilution (LID) usually was within ±1 dilution steps (ecotoxicity tests) and ±2 dilution steps (leaching and ecotoxicity tests). This is acceptable, when considering that the overall variability of sample preparation, leaching test, and bioassays add up. The conclusions lead to practical recommendations for a suitable combination of leaching and ecotoxicity tests.

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1. Introduction

The Construction Products Regulation (EU) No 305/2011 (CPR) includes the possibility to require test results to ensure that construction works are designed in such a way that they will not be a threat to the environment throughout their life cycle. Among other issues the release of dangerous substances into ground water, marine waters, surface waters or soil is taken into account in Annex 1 of the CPR, describing basic requirements for construction works. Currently, the European Committee for Standardization (CEN) within the Technical Committee 351 (CEN/TC 351) elaborates several standards for the assessment of the release of dangerous substances. Among these standards the CEN/TS 16637 part 1–3 describe the selection and performance of different leaching tests. Currently, a technical CEN guidance on the use of ecotoxicity tests applied to construction products is being developed under CEN/TC

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351.

Knowledge about potential toxic building materials and their avoidance has been identified as a key issue in sustainable construction (Pacheco-Torgal and Jalali, 2011; Ilvonen, 2013). It is generally accepted that chemical analysis of dangerous substances identified in the leachates of construction products do not cover the overall potential hazard of construction products, especially for complex matrices of unknown organic compositions. Thus the application of bioassays using leachates from complex construction has been highlighted as complimentary tool to chemical analysis by national committees (DIBt, 2009, 2011) and the issue has been picked up as a work item under CEN/TC 351. By direct ecotoxicity testing of leachates all effects of dangerous or hazardous substances to living organisms, including additive, synergistic and antagonistic effects are covered, whereas only the bioavailable fraction is taken into account. As a general rule of environmental effect analyses, different reference species, representing the relevant trophic levels of ecosystems are used for assessing the effects to a test battery.

One challenge of the ecotoxicity assessment of construction products is the selection of a suitable combination of leaching tests with ecotoxicity tests. Most studies applying such a testing strategy of combining leaching with ecotoxicity tests have been undertaken to the classification of waste to the hazard property H14 (ecotoxic waste) according to Directive 2008/98/EC (CEN/TR 16110, 2010; Stiernström et al., 2011; Weltens et al., 2012; Pandard and Römbke, 2013; Moser and Römbke, 2009). Hereby, it is generally accepted that results obtained from direct ecotoxicity testing override results obtained from H14 calculations similar to that of the Global Harmonised System (Hennebert et al., 2014).

Similar strategies have been followed for the ecotoxicological characterisation of soils, sediments or contaminated sites (ISO 15799, 2013; Feiler et al., 2013; Hafner et al., 2015). The recycling of debris from the excavation industry and drilling procedures and other byproducts for secondary construction materials for buildings and roads is another issue where the waste sector and the construction sector overlap (Stiernström et al., 2014a,b; Baderna et al., 2015).

Two technical guidance documents have been developed under CEN and ISO for suitable testing for waste and soil (CEN/TR 16110:2010; ISO 15799, 2013). In the field of construction products several studies considered the leaching of e.g. biocides from materials, some of which have been combined with ecotoxicity tests (Burkhardt et al., 2009). However, only few studies considered the ecotoxicity of complex construction products so far (Lalonde et al., 2011; Krüger et al., 2013; Sudár et al., 2013).

The aim of the project was to develop recommendations for a test battery for the ecotoxicological assessment of the environmental impact of construction products for outdoor use, which contributes to the European harmonization of test methods. The focus of the study was on possible impacts to surface water while the sediment or terrestrial compartment or emissions to the air were not covered.

2. Materials and methods

Construction products. In total 39 representative construction products were selected based on sector specific expertise, and preliminary elution tests were performed (data not shown). The focus was mainly on construction products containing leachable organic substances. Of these, 20 products were included in the ecotoxicological testing program and were eluted again. These products belong to different categories such as PUR-foam, wood-plastic-composites, sealing masses, plastic and bitumen sheets, sport floors, EPDM- and TPES-granulates as subfloor for sport fields, geo textiles resp. mat of fibres as well as different construction materials made of plastics (sewer pipe, rainwater down pipe, acryl glass, PC-twin-wall-sheet (see Table 1)). Reactive construction products were applied e.g. to glass plates according to the manufacturer instructions in terms of preparation, mixing, drying time, etc.

Leaching tests. The construction products were tested with leaching methods developed e.g. by the CEN/TC 351 working group and were adapted to the requirements of ecotoxicity testing: In total 17 monolithic and planar construction products were eluted with deionized water in the Dynamic Surface Leaching test (DSLT) according to CEN/TS 16637-2. The volume/surface area ratio was set at 20 L m^{-2} while using the mixture of the first two leaching steps after 6 and 18 additional hours.

Three granular construction products were eluted in the one stage batch test according to ISO 12457 Part 1, which was originally developed for waste samples. Here, the sample is eluted in an overhead shaker with a water/solid ratio of 2 L kg^{-1} over 24 h. In the technical specification CEN/TS 16637-1 of CEN/TC 351 the test is mentioned as “indirect method”.

Basic analytics. The eluates were subjected to a chemical-based analysis. At BAM the TOC was determined with a TOC-VCPH-Analyzer (Shimadzu, Berlin, Germany). Cations and anions were quantified by ion chromatography (Dionex 320, Dionex GmbH, Idstein, Germany) in combination with ICP/OES (iCAP 7000) or ICP/MS (ICAP Q, both Thermo Scientific, Schwerte, Germany), conductivity and pH measured with a testo® 240 (Testo GmbH, Lenzkirchen, Germany) and a pH 540 GLP (WTW GmbH, Weilheim, Germany). In addition, GC-passing substances were qualitatively identified by gas chromatography coupled with mass spectrometry (GC/MS). For this, the eluates were extracted with toluene, dichloromethane and ethyl acetate for 20 min on an orbital shaker and re-dissolved in toluene, after phase separation, drying and concentration via rotary evaporation, using gas chromatography (Agilent Technologies 6890N, Waldbronn, Germany) and mass spectrometer (Agilent Technologies 5973 Network, Waldbronn, Germany).

At Hydrotox the pH, the oxygen concentration, the conductivity of the eluates was determined again using a WTW Multi 9430 with SenTix® 940, FDO® 925, and TetraCon® 925 sensors (all WTW GmbH, Weilheim, Germany). The total organic carbon (TOC) was measured using a total carbon analyzer (TOC-5000A, Shimadzu Germany, Duisburg). The measurement principle follows catalytically aided combustion oxidation at 900 °C after purging the dissolved inorganic carbon (carbonate carbon) with oxygen.

Ecotoxicitcy tests. According to DIBt principles four aquatic tests (algae, daphnia, luminous bacteria, fish eggs), one genotoxicity test (umu test) and one test for ready biodegradability (OECD 301 F) were used. Terrestrial tests were not considered in this project. The results of ecotoxicity tests were indicated as LID (Lowest Ineffective Dilution). The LID corresponds to the lowest dilution level “D” at which no inhibition, or only effects not exceeding the test-specific variability, are observed (ISO/DIS 5667-16: 2016).

The **Algal Growth Inhibition Test** was performed according to ISO 8692 (2012). The first series of experiments was performed using the green algae *Desmodesmus subspicatus* CHODAT (SAG 86.81). For subsequent series the green algae species *Pseudokirchneriella subcapitata* (*Raphidocelis subcapitata*, SAG 61.81) was used, because the ISO validity criterion for the growth rate of controls of 1.4 day^{-1} was not always achieved with *Desmodesmus subspicatus*. For some eluates valid studies for both species were performed. In total 3 replicates per concentration and 6 control vessels of 50 mL each were incubated in a temperature controlled light incubator (RUMED 1301, Laatzen, Germany). Temperature was 23 °C ± 1 °C, light intensity was in the range between 111 and

Table 1
Construction products considered in the study.

Product		Standard	Identified substances in eluates
BAM-1	Polyurethane foam	EN 15651-1	Triphenyl phosphate TCPP, N-Phenylformamide Nickel 30 μg L^{-1}
BAM-2	Acrylic sealant	EN 15651-1	Isobenzofuranone Phthalates Antimony 9 μg L^{-1}
BAM-4	Acrylic resin sealant		Phthalate Zinc 95 μg L^{-1}
BAM-5	PVC-U pipe	prEN 13476 and EN 15012	*GC/MS signals and mass spectra do not meet identification criteria*
BAM-6	Plastic pipe	EN 15012	*GC/MS signals and mass spectra do not meet identification criteria*
BAM-8	Acrylic glass	EN 1013	*GC/MS signals and mass spectra do not meet identification criteria*
BAM-10	PVC panel		*GC/MS signals and mass spectra do not meet identification criteria*
BAM-11	Wood-Plastic-Composite (WPC)	EN 15534-1	Vanillin Boron 46 mg L^{-1} Copper 12 μg L^{-1}
BAM-12	Polycarbonate twin-wall sheet	EN 16153	*GC/MS signals and mass spectra do not meet identification criteria*
BAM-13	Sealing mass		Diethyl phthalate, 2-Ethylhexyl Butyrate 1(3H)-Isobenzofuranone Nickel 74 μg L^{-1}
BAM-14	Sports floor made of ethylene propylene diene monomer (outdoor use)	EN 14877	Mercaptobenzothiazoles & degradation products Thiourea derivatives, Phenole derivatives Zinc 173 μg L^{-1}
BAM-22	Bitumen thick film	EN 14023	Phthalates Zinc 299 μg L^{-1}
BAM-G1	EPDM resin, made of ethylene propylene diene monomer	granular raw material used as subfloor	Trimethylthiourea, Thiourea derivatives Mercaptobenzothiazoles & degradation products Zinc 191 μg L^{-1}
BAM-G2	TPES resin, made of thermoplastic elastomers (styrenic block copolymers)	granular raw material used as subfloor	2,2,6,6-Tetramethyl-4-piperidone 2(3H)-Benzothiazolone, 2-Mercaptobenzothiazol Vanadium 7.4 μg L^{-1} Zinc 7,4 μg L^{-1}
HSR-2	EPDM (roof covering panel)		Zinc 150 μg L^{-1} Benzothiazole
			Expected substances in eluates
HSR-3	FKS liquid synthetic sealant (roof covering)		Polymethylmetacrylate (PMMA), amine, isocyanate
HSR-6	PVC panel (roof covering)	EN 13956	Plasticizers (911-phthalate, DINP, DIDP), Isodecylphenylphosphite, Sn-compounds
HSR-7	Elastomer bitumen sealant panel		Root protection agent (ester of Mecoprop)
HSR-10	PET Multifil with polymer covering	Geotextile	Antimony, plasticizers
HSR-12	Core: polyamid monofilament, coating: PP fleece	Geocomposite	Antimony, plasticizers

EN 1013 (2015) Light transmitting single skin profiled plastics sheets for internal and external roofs, walls and ceilings.
pr EN 13476-1 (2015) Plastics piping systems for non-pressure underground drainage and sewerage - Structured-wall piping systems of unplasticized poly(vinyl chloride) (PVC-U), polypropylene (PP) and polyethylene (PE) — Part 1: General requirements and performance characteristics.
EN 13956 (2012) Flexible sheets for waterproofing - Plastic and rubber sheets for roof waterproofing - Definitions and characteristics.
EN 14023 (2010) Bitumen and bituminous binders - Specification framework for polymer modified bitumens.
EN 14877 (2013) Synthetic surfaces for outdoor sports areas - Specification.
EN 15012 (2008) Plastics piping systems - Soil and waste discharge systems within the building structure - Performance characteristics for pipes, fittings and their joints.
EN 15534-1 (2014) Composites made from cellulose-based materials and thermoplastics (usually called wood-polymer composites (WPC) or natural fibre composites (NFC) — Part 1: Test methods for characterisation of compounds and products.
EN 15651-1 (2012) Sealants for non-structural use in joints in buildings and pedestrian walkways — Part 1: Sealants for facade elements.
EN 16153 (2015) Light transmitting flat multiwall polycarbonate (PC) sheets for internal and external use in roofs, walls and ceilings - Requirements and test methods.

118 μE m^{-2} s^{-1} varying not more than 10% between the incubation levels. The algae chlorophyll-fluorescence after an exposure time of 72 h was measured after transforming samples into a 96-well micro-plate, using a fluorescence micro-plate reader (Tecan infinite F200, Männedorf, Switzerland, excitation wave length: 465 nm, emission wave length: 670 nm). The correlation factor between chlorophyll-fluorescence and algae cell count was determined using a Coulter Counter Z2 (Beckman Coulter, Krefeld, Germany).

The **Daphnia Acute Toxicity Test** was performed according to EN ISO 6341 (2013) with *Daphnia magna*. The Daphnids were held in M4 medium and were fed every working day with an algal suspension of *Desmodesmus subspicatus* ensuring a carbon (C) feeding of 0.1 mg C per animal and day. Each 2 beakers per concentration with 5 daphnids were exposed at 20 ± 1 °C and a light-dark rhythm of 16:8 h. Daphnids were examined after 24 h and 48 h.

The **Fish Egg Test** was performed according to ISO 15088 (2007) with immediate fertilized eggs from *Danio rerio*. Incubation was performed in polystyrene 24-well microplates with a sample volume of 2 mL at 26 °C ±1 °C. The test design corresponded to 10 replicates (wells) per concentration, four internal negative controls and negative controls and positive controls. Embryos were examined after 24 h and after 48 h. The standard visual mortality criteria of the test guideline (coagulated eggs, no tail detachment, and non-detectable heartbeat) were applied. Other observations such as "no somites" were reported, but not included in the evaluation.

The **Luminescent Bacteria Test** was performed according to ISO 11348-2 (2007) with *Vibrio fischeri* obtained as freeze dried bacteria from Hach Lange GmbH, Düsseldorf (LCK 482) while using two replicates per concentration. Incubation period was 30 min at 15 ± 1 °C varying not more than 0.5% between incubation places.

The **umu Test**, a genotoxicity test with the bacterial strain *Salmonella typhimurium TA1535/pSK1002,* was performed according to ISO 13829 (2000). The bacteria are exposed for 2 h to the eluate with and without metabolic activation followed by a growth phase of 2 h, and the genotoxin-dependent induction of the umuC-gene was compared to the spontaneous activation of the control culture. Each concentration has been tested three-fold in 96-well microtitre plates (Greiner Bio-One, Frickenhausen, Germany). The induction rate (IR) corresponds to the increase of the extinction at 420 nm relative to the negative control. Bacterial growth and inhibition are determined turbidimetrically from the optical density at 600 nm. For growth factors below 0.5 (50% growth inhibition) the results are not evaluated. The result given is the smallest dilution step at which an induction rate <1.5 is measured. Samples which are toxic at higher concentrations but non-genotoxic at growth factors >0.5 were designated as "toxic".

Biodegradability. Eluates whose TOC exceeded ca. 10 mg L^{-1} were examined in the respirometer test according to OECD 301 F using the OxiTop® tests system (WTW, Weilheim, Germany). This consists of narrow-necked glass bottles with rubber sleeve inserts for NaOH pellets, in which the carbon dioxide evolved is absorbed. In total three reactors containing the eluates, three reactors containing only inoculum (blank), and three reactors containing sodium acetate as reference compound were positioned on a stirrer platform. The liquid volume was fixed as 164 mL each. The activated sludge inoculum (final concentration 30 mg L^{-1} dry solids) was freshly derived from the municipal wastewater treatment plant Breisgauer Bucht (600.000 inhabitant equivalents). The bottles were sealed tightly with the measuring heads and every 112 min the pressure was measured for each bottle and stored. The chemical oxygen demand (COD) of the eluates was determined colorimetrically using vial test cuvettes (LCK 114, LCK 314) with a CADAS 200 photometer (Hach Lange GmbH, Düsseldorf, Germany) and was adjusted to the test concentration of 50–100 mg L^{-1}. The test duration was 28 days, the extent of biodegradation is indicated as % COD.

3. Results

Chemical characteristics of eluates. The maximum TOC values were obtained with sealants BAM-2 and BAM-4 (up to 460 mg L^{-1} TOC) followed by the wood-plastic composite BAM-11 and EPDM materials HSR-2 and BAM-G1. The pH varied between 4.2 (PU foam BAM-1) and 8.1 (plastic pipes), the electric conductivity between 2 and 345 µS cm^{-1} (Table 2).

Ecotoxicity of eluates. Four eluates (BAM-14, BAM-G1, HSR-2, HSR-3) demonstrated very high ecotoxicity in several tests (Table 3). The maximum LID values were observed with the BAM-G1 eluate (luminescent bacteria $LID_{lb} = 1536$, algae $LID_A = 192$, daphnia $LID_D = 192$). With a few exceptions, the daphnia and fish egg tests showed a significantly lower toxicity. The highest fish egg toxicity was observed with the acrylic sealant BAM-4. ($LID_{Egg} = 12$). When comparative data were available, the algae species showed a similar sensitivity. None of the samples tested was genotoxic in the umu test.

The reproducibility of the test results obtained with the eluates in test repetitions with overlapping concentrations was in most samples well (±1 dilution step, data not shown). In the three blank controls in the DSL test and one stage batch leaching test little or no toxicity was observed. This confirms that the materials used for the leaching tests do not induce significant effects that could wrongly be attributed to the construction products.

Biodegradability. The samples showed mainly a good biodegradability of 75%–97% (BAM-2, BAM-4, BAM-11, BAM-22, BAM-G1, HSR-12). In contrast, the eluate of BAM-G2 only reached a biodegradation of 44%, the eluate of HSR-3 a biodegradation of 46% (Figs. 1 and 2).

Intra-laboratory repeatability. To confirm the final selection of the construction products for the round robin test, 5 construction products were again eluted in the DSL test or one stage batch test and were then examined in the ecotoxicity tests. Hereby, also the reproducibility and robustness of the overall process obtained in one laboratory could be verified. It was demonstrated that the TOC can vary by a factor of 1.5–3. In particular, the hardening construction products HSR-3 (factor 2.6) and BAM-22 (factor 1.7) were outstanding (Table 4). The test results of the ecotoxicity tests were

Table 2
Chemical characterisation of eluates.

Product		Elution	TOC mg L^{-1}	COD mg L^{-1}	pH	Conductivity µS cm^{-1}
BAM-1	Polyurethane foam	DSL	28.6	43.4	4.2	21
BAM-2	Acrylic sealant	DSL	469.1	1549	6.2	280
BAM-4	Acrylic resin sealant	DSL	250.2	1062	4.7	10.3
BAM-5	PVC-U pipe	DSL	1.1	–	8.1	10.9
BAM-6	Plastic pipe	DSL	1.7	–	8.1	15.9
BAM-8	Acrylic glass	DSL	1.4	–	8.1	5.2
BAM-10	PVC panel	DSL	1.4	–	7.6	5.2
BAM-11	Wood-Plastic-Composite (WPC)	DSL	68.3	–	7.9	345
BAM-12	Polycarbonate twin-wall sheet	DSL	0.6	–	7.7	1.6
BAM-13	Sealing mass	DSL	6.1	–	5.8	17.8
BAM-14	Sports floor	DSL	7.3	–	5.1	24.6
BAM-22	Bitumen thick film	DSL	44.4	136	6.3	323
BAM-G1	EPDM resin	Batch	61.2	82.3	6.0	208
BAM-G2	TPES resin	Batch	29.9	71.5	6.0	202
Hydrotox-DSLT	Blank DSLT	DSL	1.2	–	8.2	2.4
BAM-DSLT	Blank DSLT	DSL	0.6	–	5.8	8.4
BAM-batch test	Blank batch test	Batch	1.0	–	5.1	12.2
HSR-2	EPDM (roof covering panel)	DSL	9.0	–	7.1	17.9
HSR-3	FKS liquid synthetic sealant (roof covering)	DSL	95.3	310	4.4	31.5
HSR-6	PVC panel (roof covering)	DSL	2.3	–	6.6	5,6
HSR-7	Elastomer bitumen sealant panel	DSL	1.9	–	6.8	19.6
HSR-10	PET Multifil with polymer covering (geotextile)	DSL	3.2	–	6.4	18.8
HSR-12	Core: polyamid monofilament, coating: PP fleece (geocomposite)	DSL	35.4	137	6.8	17.4

Table 3
Screening test results with ecotoxicity tests.

Product		Algae		Daphnia	Fish	Bacteria	umu
		LID_A Desmo-desmus	LID_A Raphido-Celis	LID_D	LID_{Egg}	LID_{lb}	LID_{EU}
BAM-1	Polyurethane foam	–	32	1	1	≤2	1.5
BAM-2	Acrylic sealant	–	32	3	1	4	1.5
BAM-4	Acrylic resin sealant	–	24	2	12	16	1.5
BAM-5	PVC-U pipe	≤2	–	≤2	≤2	≤2	1.5
BAM-6	Plastic pipe	≤2	–	≤2	≤2	≤2	1.5
BAM-8	Acrylic glass	≤2	–	≤2	≤2	≤2	1.5
BAM-10	PVC panel	≤2	–	≤2	≤2	≤2	1.5
BAM-11	Wood-Plastic-Composite (WPC)	3	–	2	≤2	≤2	1.5
BAM-12	Polycarbonate twin-wall sheet	≤2	–	≤2	≤2	≤2	1.5
BAM-13	Sealing mass	–	8	1	1	6	1.5
BAM-14	Sports floor	–	64	24	3	192	1.5
BAM-22	Bitumen thick film	–	4	2	6	6	1.5
BAM-G1	EPDM resin	–	192	192	4	1536	1.5
BAM-G2	TPES resin	–	16	1	1	64	1.5
Hydrotox-DSLT	Blank DSLT	≤2	–	≤2	≤2	≤2	1.5
BAM-DSLT	Blank DSLT	–	>1	1	1	≤2	1.5
BAM-batch test	Blank batch test	–	>1	1	1	≤2	1.5
HSR-2	EPDM (roof covering panel)	12	16	4	1	128	1.5
HSR-3	FKS liquid synthetic sealant (roof covering)	>48	512	3	3	12	1.5
HSR-6	PVC panel (roof covering)	1	1	1	1	≤2	1.5
HSR-7	Elastomer bitumen sealant panel	1	1	1	1	≤2	1.5
HSR-10	PET Multifil with polymer covering (geotextile)	1	4	2	1	≤2	1.5
HSR-12	Core: polyamid monofilament, coating: PP fleece (geocomposite)	≤2	3	4	2	8	1.5

within the expected range. The deviation of the LID in both testing series was usually ±2 dilution steps. This is considered acceptable, because the variability of sample preparation, washout, and bioassays add up (Table 5). One exception again was BAM-22 whose eluates showed differing results, probably due to different hardening conditions of test specimens.

4. Discussion and recommendations

Direct testing of eluates from construction products is a very promising approach to assess the release of dangerous substances from construction products into the environment. Here, the combined effects of all leachable substances and transformation products are determined over their total effects in bioassays. The focus is on construction products whose ingredients are only detectable by analytical methods, if any, with a high effort. By means of the ecotoxicological characterisation also a prioritization of construction products to be submitted for a comprehensive evaluation (including chemical analysis) can be carried out. Ecotoxicity testing is a complementary tool to chemical analysis suitable for certain product groups in combination to this.

Preparation of test specimens. Representative test samples were prepared according to the manufacture instructions. However, sample preparation proved to be critical, especially for hardening or reactive construction products. In our study a significant part of the variability observed was attributed to the test samples. Another issue is cut edges of specimens that do not come into contact with water in the usual application and which should be sealed with a toxicologically acceptable material before performing the elution test. The actual surface area cannot be determined for construction products with e.g. irregular and lattice-like surfaces.

Selection of leaching tests. The agreement to use the first two fractions of the DSL test for the ecotoxicological tests leads to the consequence that substances may be washed off from the surface of the specimen, thus causing relatively high release rates. Later on, the leaching process for many construction products is controlled by diffusion of substances to the material surface and the washout rate is reduced over time. For some products, the very long contact times in the last leaching phases of the DSL-test may also cause higher concentrations. A long contact time may also have the disadvantage that (bio) degradable ingredients are not gathered.

For the one stage batch tests for granular construction products the water contact is far more intense than for DSLT. Similar leaching tests with a water/solid ratio of 10 L/kg are also widely used and suitable. Shaking is a practical, simple process, and relatively high concentrations in the leachate can be expected at a low water/solid ratio. The specific ratio of the volume of water to the surface (DSLT) or the weight (shaking test) has a decisive influence on the comparability of test results and should be clearly defined. The determination of the test fractions to be tested depends on the objective.

The column test according to CEN/TS 16637 part 3 was not considered in our study because at the time when starting our investigation no agreement about the boundary conditions of the test was obtained. This test may provide more realistic long term test conditions for granulates and should be a focus of future research.

Biotest battery. The approach applied in the project considers a combination of elution tests for construction products with a test battery representing different trophic levels (algae as primary producers, daphnia as primary consumers, fish eggs as secondary consumers and bacteria as decomposers). The study showed significantly differing ecotoxicity patterns of eluates from construction products in the different bioassays. Basically, the various biological effects can only be represented by a test battery and not by a single organism. For practical reasons the tests used can be allocated to short-term tests (acute toxicity). Chronic toxicity testing would be associated with much higher costs. It is recommended to use only standardized and validated test methods (EN ISO or OECD Guidelines).

The intra-laboratory variability of the ecotoxicity tests at the Hydrotox laboratory usually was in the range ±1 dilution levels (assuming a factor of 2 in the concentration). In subsequent repetitions of the leaching and ecotoxicity tests with 5 construction products both the chemical sum parameters (TOC, conductivity) as well as the ecotoxicity showed an acceptable reproducibility in the

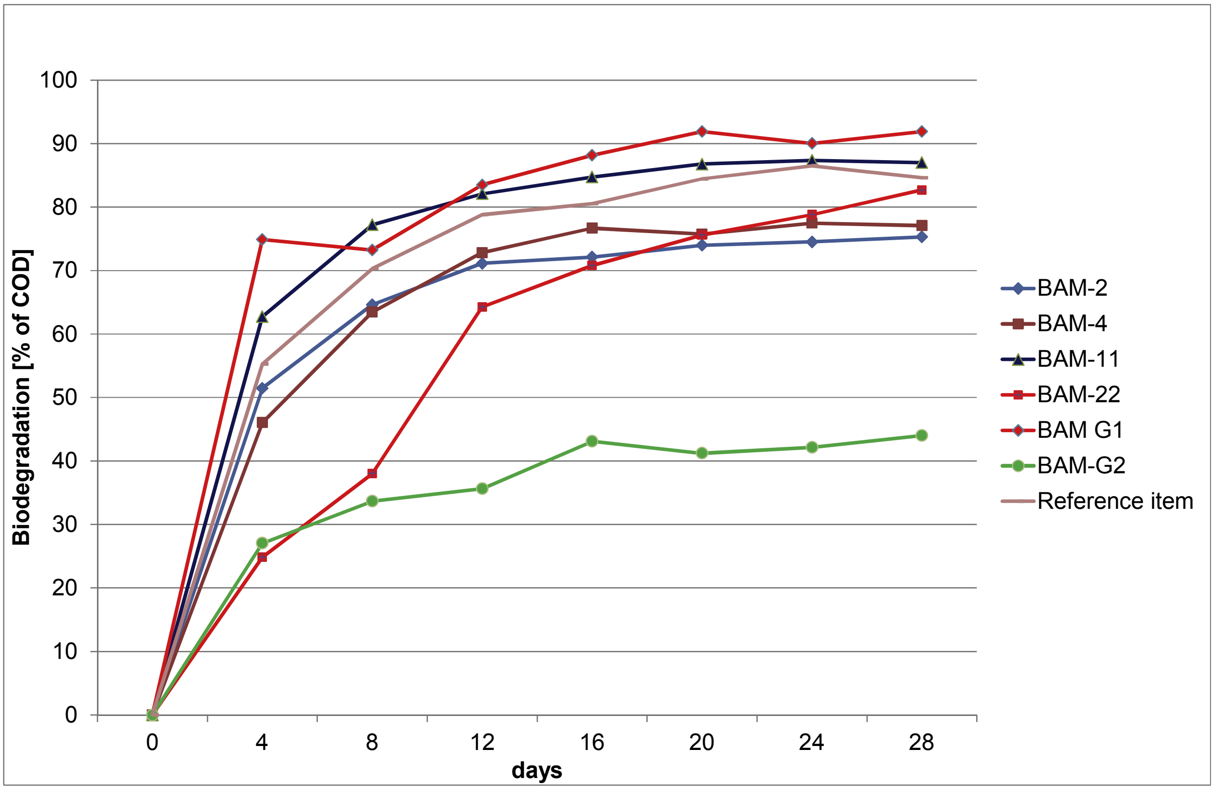

Fig. 1. Biodegradability of BAM samples in the Respirometer test.

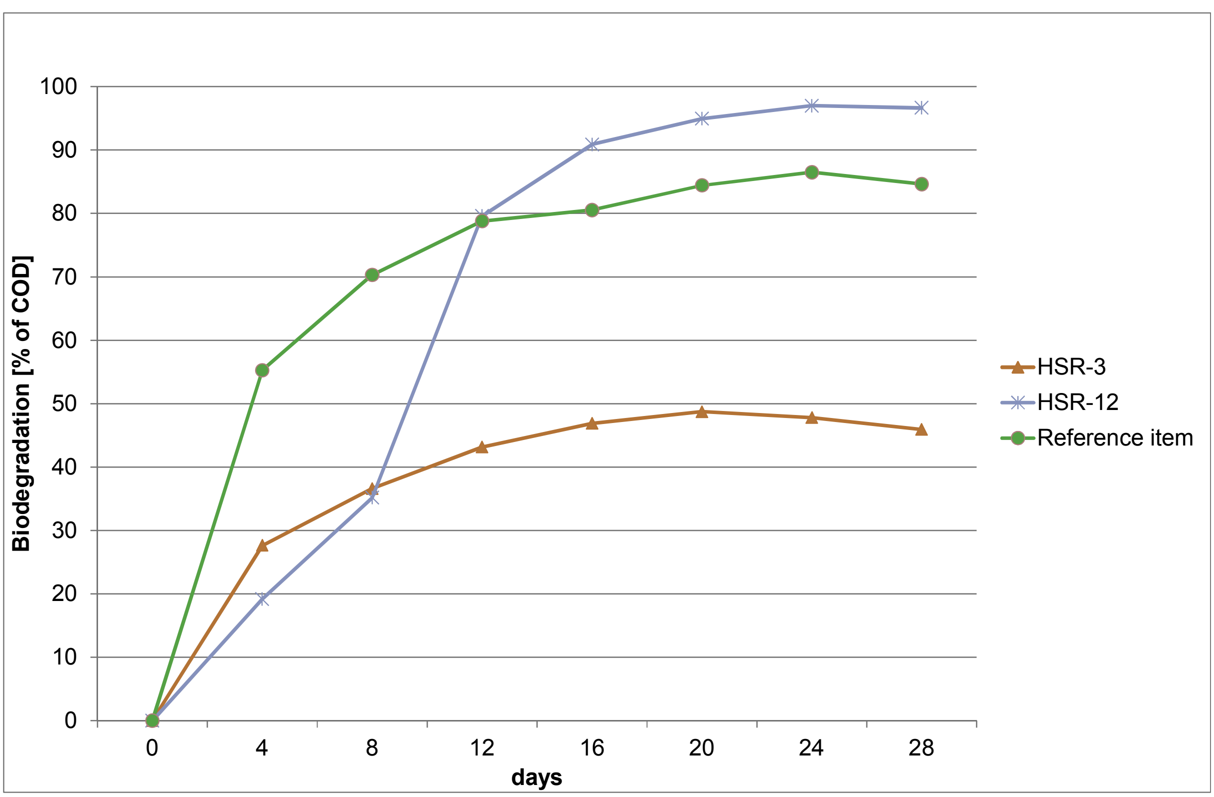

Fig. 2. Biodegradability of HSR samples in the Respirometer test.

Table 4
Intra-laboratory repeatability of chemical parameters.

Year	Product	2013		2014	
		TOC mg L^{-1}	Conductivity µS cm^{-1}	TOC mg^{-1}	Conductivity µS cm^{-1}
HSR-2	EPDM panel	9.0	17.9	5.2	13.9
HSR-3	FKS liquid sealant	95.3	31.5	36.3	22.9
BAM-22	Bitumen thick film	44.4	323	25.4	247
BAM-G1	EPDM resin	61.2	208	46.4	187
BAM-G2	TPES resin	29.9	202	24.1	213
Blank	DSLT	1.2	2.4	–	5.4
Blank	Batch test	1.0	12.2	0.2	4.0

Table 5
Intra-laboratory repeatability of ecotoxicity tests.

Year	Product	Algae		Daphnia (48 h)		Fish egg		Bacteria	
		2013	2014	2013	2014	2013	2014	2013	2014
		LID_A	LID_A	LID_D	LID_D	LID_{Egg}	LID_{Egg}	LID_{lb}	LID_{lb}
HSR-2	EPDM panel	12	6	4	≤2	1	1	128	64
HSR-3	FKS liquid sealant	512	≤192	3	4	3	2	12	8
BAM-22	Bitumen thick film	4	>6	2	16	6	12	6	8
BAM-G1	EPDM resin	192	192	192	384	4	6	1536	1024
BAM-G2	TPES resin	16	4	1	2	1	1	64	48
Blank	DSLT	>1	1	1	2	1	1	≤2	≤2
Blank	Batch test	>1	≤2	1	≤2	1	1	≤2	≤2

range of ±2 dilution levels. Exceptions include sealants or reactive or hardening construction products.

Consideration of blank controls. The consideration of blank controls over the whole process, from the leaching test, the pretreatment and storage of eluates, and the ecotoxicity tests is recommended. In our study these blank controls showed little or no toxicity, but these additional controls are suitable to detect any contamination of the equipment or dilution water which might be wrongly attributed to the construction products.

Origin of ecotoxicity. Although the identification of ingredients causing ecotoxic effects was not a primary objective of the study, the focus being on the suitability of overall approach, some conclusions can be drawn from literature. The BAM-G1 and HSR-2 construction products, which later have been selected for the round robin test (Gartiser et al., 2016), both consist of ethylene propylene diene monomer (EPDM). Among the organic substances identified in the eluates are trimethylthiourea and derivatives, mercaptobenzothiazoles and degradation products as well as zinc (>100 µg L^{-1}). The toxicity of zinc is attributed to the water soluble form and depends on the pH, water hardness, DOC etc. In the European Risk Assessment Report for Zinc the EC50 (72 h) with the algae *Pseudomonas subcapitata* was within the range of 25.8 and 1630 µg Zn L^{-1} and the EC10 (72) between 4.8 and 608 µg Zn L^{-1}. The acute EC50 for *Daphnia magna* was reported to be in the range of 70–860 µg Zn L^{-1} (EU, 2010). When comparing with the measured zinc concentrations (Table 1) the zinc toxicity could therefore explain some, but not all of the ecotoxicity observed in the eluates. The ecotoxicity of 2-mercaptobenzthiazole also identified in the eluates is in the range of EC50 (72 h) = 0.5 mg L^{-1} (algae) resp. EC50 (48 h) = 0.71 mg L^{-1} (ECHA, 2016). While for 2-mercaptobenzthiazole a limited biodegradation in OECD 301 tests has been reported, in our study the EPDM eluates as a complex mixture showed considerable biodegradation in the OECD 301 F test, indicating, that 2-mercaptobenzthiazole is not the primary organic ingredient in the leachates. Other studies confirmed the high number of hazardous substances released from rubber material (Llompart et al., 2013).

Krüger et al. (2013) studied the ecotoxicity behavior of leachates from granular sport ground materials using a similar batch test according to DIN 19529 with a liquid to solid ratio of 2 L kg^{-1}. The highest effects on *Daphnia magna* (EC50 < 0.4% leachate) were observed with foamed EPDM while the highest algal toxicity (EC50 = 15.6% leachate) was detected with styrene butadiene rubber (SBR) leachates. The toxicity of the investigated thermoplastic elastomer was relatively low towards algae and moderate towards daphnia compared to the other materials. No correlations between ecotoxicity and zinc and PAH concentrations were observed. Comparative analysis with leachates obtained from column tests, suggested to better reproduce field conditions, generally showed lower or no ecotoxic effects compared to eluates obtained from batch tests. Wik and Dave (2006) studied the toxicity of leachates from tier wear rubber rasps, exposed 72 h at 44 °C, against *Daphnia magna* and found EC50 (48 h) values ranged from 0.5 to >10.0 g L^{-1}. The analysis revealed that non-polar organic compounds caused most of the toxicity.

The difficulty to assign the contribution of particular substances to the overall ecotoxicity of eluates from construction products on the other hand demonstrates the benefit from direct ecotoxicity testing of such complex mixtures as a supplementary evaluation tool next to chemical analysis.

Regulatory evaluation. On a European level technical guidance on the use of ecotoxicity tests for the evaluation of construction products is currently being developed in the European Committee for Standardization in the Technical Committee "Construction Products: Assessment of the Release of Dangerous Substances" (CEN TC 351). On a national level, Germany applies a testing scheme with selected aquatic tests, also applied in the project, for which moderate aquatic ecotoxic effects (LID 4-8), but no genotoxicity is considered as an indicator that the requirements are fulfilled (DIBt, 2009). When defining limit values, the different leaching conditions should carefully be considered.

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