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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.

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