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# ECOTOXICOLOGICAL ASSESSMENT OF RECYCLED AGGREGATE AND CONCRETE VIA AQUATIC BIOTESTS

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Abstract. With the growing consumption of primary raw materials, the need for recycling of construction and demolition waste (CDW) arises. According to international and national regulations, any waste must be tested for ecotoxicity using a leaching test followed by chemical analysis and a set of aquatic toxicity biotests. Standardized leaching procedures have also been developed for construction materials and products and are used in common practice. On the other hand, studies aimed at direct determination of ecotoxicity are still lacking. Acute toxicity tests with unicellular algae, freshwater crustaceans, and marine bacteria are among the most popular for determining the ecotoxicological potential of recycled aggregates or concrete samples. The article deals with the perspective of semichronic and chronic tests with extended exposure, as well as testing of leachates obtained from leaching events for more than 24 hours. Results of performed experiments were compared and evaluated. From the point of view of sustainability, it is necessary to develop an optimal experimental design for the ecotoxicological evaluation of recycled aggregate and concrete. The conclusion of the paper is the evaluation of possible methods and their combinations.

Keywords: Recycled aggregate, recycled concrete, construction and demolition waste (CDW), leachate, ecotoxicity.

## 1. Recycling of CDW – A step toward sustainability

The construction sector is one of the largest consumers of mineral resources, which is also related to the subsequent production of waste. The production of the most widely used products in this sector, such as bricks and concrete, is currently dependent on the constant extraction of primary materials from nonrenewable resources [1]. Reuse of recycled materials in the construction industry has considerable potential for saving primary resources [2–4]. At the same time, recycling is also beneficial due to the reduction of construction and demolition waste and its disposal in landfills. The life cycle of construction materials is shown in Figure 1.

In addition to the importance of the mechanical and chemical characteristics of the materials, mainly recycled aggregates, it is appropriate to assess the degree of impact on the environment. Recently, many studies have relied on life cycle assessment (LCA). During the recycling process, the CDW undergoes several steps of conversion to secondary raw material. Efforts have been made to facilitate this process for several years, for example, by replacing classic demolitions with so-called controlled demolitions [5, 6]. During this process, the object (structure) is disassembled and sorted according to individual materials, to facilitate preparation for the next process, reuse, and recycle. This attitude is quite common, e.g. in Belgium.

However, due to different international and/or national regulations, different criteria may apply to waste and different to secondary raw materials (recycled aggregates). The problem is generally the testing of the physical and mechanical properties of these materials, because although they are mainly defined as waste, in the case of recycled aggregates or recycled concrete, tests are necessary according to the standards for aggregates and concrete mixtures [7]. In terms of further use, it is possible to apply modified CDW in construction. However, when dealing with CDW, the legislation is highly complicated. A problematic question arises in case that the CDW is classified as hazardous.

## 2. Leaching tests

Generally, there are two ways to characterize construction materials or wastes from a chemical and/or environmental point of view. Analysis of the samples in solid form using X-ray based methods brings insight into chemical composition and reveals potentially hazardous substances. However, information on chemical concentration does not fully refer to the bioavailability of the compounds and thus the level of (eco)toxicity. Another option is to perform leaching tests and subsequent chemical analysis of both inorganic and selected organic compounds in the leachate. The leaching

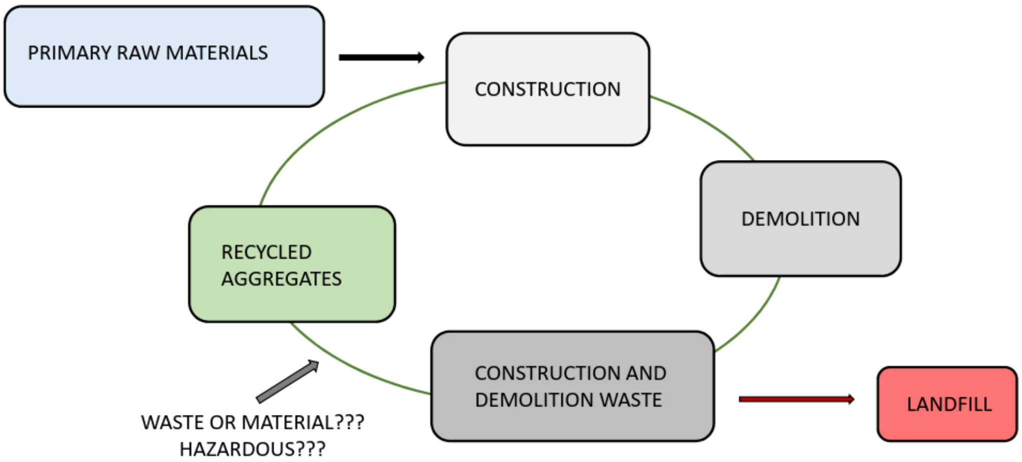


Figure 1. Life cycle and recycling possibilities of primary raw materials.

| Method | Experimental design | Duration | Leachant renewal | Reference |
|---|---|---|---|---|
| EN 12457-4 | One-stage batch; homogenized sample | 24 h | No | [8] |
| EN 16637-3 | Percolation; granular material | 24 h | No | [9] |
| CEN/TS 15862 | One-stage batch; monolith | 24 h | No | [10] |
| EN 16637-2 | Multiple-stage | 64 d in total; (stages 6 h – 28 d) | Yes | [11] |

Table 1. Leaching standards used for characterization of construction materials, products, and wastes.

methods mainly applied for the characterization of construction products, wastes and recycled aggregates are presented in Table 1. One-stage batch leaching of homogenized material under continuous shaking [8], a method determined for wastes can also be chosen for the testing of recycled aggregates [12–15] or crushed concrete products [13, 16, 17].

Using a one-stage batch or long-term multi-stage leaching procedure without the homogenization step [10, 11] represents a more suitable approach for testing construction products, such as concrete samples. In contrast with the EN 12457-4 [8] where leachant volume is calculated according to sample weight, in case of both CEN/TS 15862 [10] and EN 16637-2 [11], the liquid amount addition is dependent on the solid sample surface area. The one-stage batch monolith leaching protocol was applied in a study dealing with slag cement-based concrete [18]. The multistage leaching method provides a more complex overview of the leaching potential of the material because both prolonged time and repeated leaching are taken into consideration. Recently, an interlaboratory test was performed using various types of samples with both a percolation test and a dynamic surface leaching test [19].

A different leaching procedure was used in our previous study [14] to compare the leaching of glass waste before and after its use in recycled high performance concrete (HPC). For both raw materials, i.e., crushed glass waste and concrete products in the form of cubes, the same liquid/solid ratio and leaching time were applied to assess the immobilization of hazardous substances in the HPC.

## 3. Ecotoxicological tests of leachates

Unlike the calculation of the ecotoxicological impact on both freshwater and marine aquatic ecosystems using the LCA methodology, a direct ecotoxicological assessment can be conducted with leachates. The principle of ecotoxicological bioassays is to expose a model organism to a set of dilutions of a tested sample (leachate) together with a negative control (culture medium). Culture medium also serves as a dilution water. The leachates can be alternatively treated such as pH adjustment in the case of extreme pH values unbearable for the survival of the model organism and/or nutrient addition. Selected test organisms are exposed to the sample sets under given conditions (duration, temperature, and light regime). At the end of the exposure, the ecotoxicological endpoint (generally survival, growth, or reproduction of the organisms) is determined. The tested organisms are compared to the control organisms. In case of statistically significant decrease/increase of the endpoint tested compared to

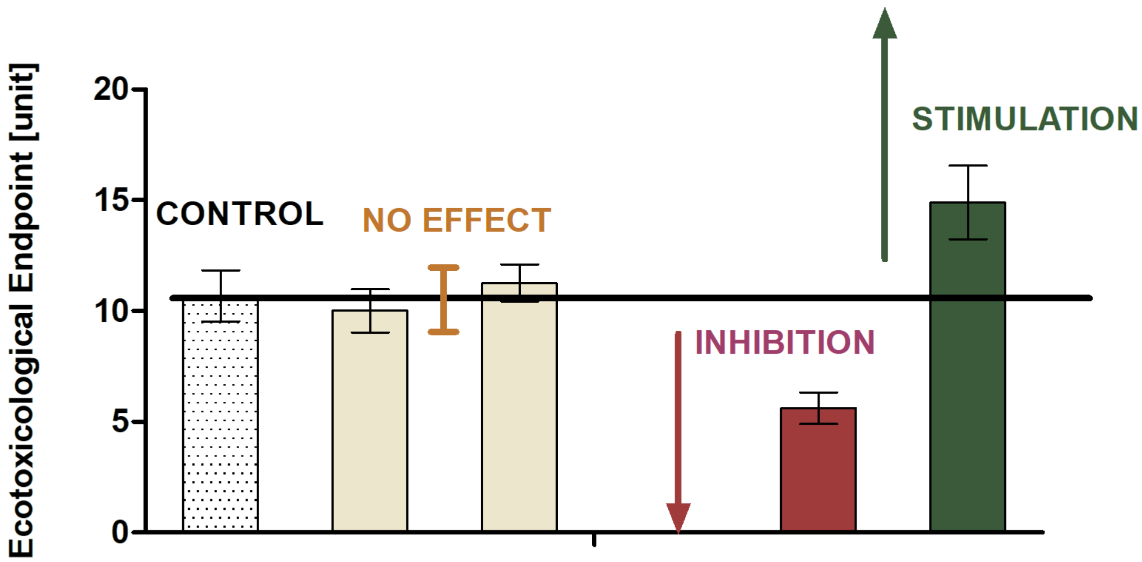


FIGURE 2. Inhibition/stimulation effect in ecotoxicological biotests.

the control, the inhibition/stimulation effect is considered, as shown in Figure 2. The inhibition/stimulation effect is usually expressed in percentage and serves for further determination of ecotoxicological indexes and evaluation of ecotoxicity risk.

For ecotoxicological assessment, it is common to select a battery of biotests which consists of organisms of all basic trophic levels, i.e. producers, consumers, and decomposers. Freshwater and marine organisms can be used. The ecotoxicological evaluation of construction samples is usually based on acute toxicity tests, which are easier to perform and last from several minutes to several days. A list of ecotoxicity tests used in studies for the construction sector is shown in Table 2.

As Table 2 shows, the most popular are acute toxicity tests with unicellular freshwater algae, freshwater crustacean, and marine luminescent bacteria. Application of acute exposure is appropriate mainly in samples with high toxic potential, where acute toxic effects such as loss of locomotion, decreased growth, and mortality are observed. With a lower concentration of potentially toxic compounds, biotests with prolonged exposure, i.e. chronic, represent a more suitable choice. However, application of such tests may be problematic not only due to the time demands, but also to the increased sample volume and space. The semichronic toxicity test with aquatic plant duckweed (*Lemna minor*) allows the determination of acute effects (necrosis, chlorosis) and chronic effects (inhibition of reproduction) with relatively short exposure time (7 days) and sample volume 100 mL per replicate [24]. At the same time, more parameters, such as growth and photosynthetic pigment content, can be determined at the morphological and biochemical levels, respectively. In contrast to unicellular algae, macroscopic plants can be exposed to unsterile samples.

Selected results from *L. minor* biotests with fine-recycled aggregates [12] and concrete mixes containing these aggregates [20] are presented in Figure 3. The photos of the duckweed plants grown for 7 days in 80 % leachates show clearly, that natural aggregate and masonry-derived recycled aggregate had no effect on plant growth, while concrete-derived recycled aggregate caused growth inhibition. However, no significant differences were observed when leachates of concrete mixes were tested.

Another option is to compare the effect of short- and long-term leaching using the same battery of bioassays. In the interlaboratory study, a series of biotests was performed with samples obtained from only 6 and 24 hours of leaching [19]. As the concentration of leached chemicals increases with time, the next step in the investigation may be to test samples obtained after several days of the leaching procedure.

It is quite clear that the ecotoxicity of concrete is dependent on the porosity (i.e. surface area) of the solid samples and chemical composition of concrete eluates. Up to date there is still limited number of studies on ecotoxicity of concrete eluates and the majority of them deals with crushed concrete. For instance, replacement of Portland cement with fly ash and natural aggregates with recycled aggregates increased the ecotoxicity of concrete eluates using particle size below 10 mm [13]. Another study on utilization of slag as fine aggregate showed, that with increasing slag amount in concrete, the leachate pH and ecotoxicity increased as well. However, the authors did not consider the level of toxicity as significant for aquatic environment [16] and similarly, only insignificant ecotoxic impact of slag cement-based concrete was determined when monolith leaching protocol was used [18]. When toxic particles are solidified in concrete mixes with small surface area, the leaching potential of the concrete is significantly reduced and thus reduced bioavailability of harmful

| | Model organism (group) | Test type | Duration | Endpoint | Reference |
|---|---|---|---|---|---|
| Producers | Freshwater unicellular algae<br>• *D. subspicatus*<br>• *R. subcapitata*[1] | acute | 72 h | growth inhibition/ stimulation | [12, 14, 15, 17, 19, 20] [21] |
| | Marine unicellular algae<br>• *P. tricornutum* | acute | 72 h | growth inhibition/ stimulation | [21] |
| | Terrestrial higher plants<br>• *Sinapis alba* | acute | 72 h | root growth | [12, 15, 16] |
| | Freshwater higher plants<br>• *Lemna minor* | semi-chronic | 168 h | growth inhibition; chlorophyll | [12, 14, 17, 20] |
| Consumers | Freshwater aquatic crustacean<br>• *Daphnia magna* | acute | 24 h; 48 h | immobilization | [12–15, 17, 19–23] |
| | Marine aquatic crustacean<br>• *Artemia franciscana* | acute | 24 h | immobilization | [21] |
| | Freshwater fish eggs<br>• *Danio rerio* | acute | 24 h; 48 h | coagulated eggs; no heartbeat, no tail detachement | [19] |
| Decomposers | Marine bacteria<br>• *Aliivibrio fischeri*[2] | acute | 15 min; 30 min | inhibition of bioluminescence | [13, 19, 21, 23] |
| | • Yeast (*S. cerevisiae*) | acute | 16 h | growth inhibition | [13, 23] |
| | Intestinal bacteria<br>• *Salmonella typhimurium* | genotoxicity | 2 h | *umuC* gene activation | [19] |

[1] *Raphidocoelis subcapitata*: former *Pseudokircheniella subcapitata*.
[2] *Aliivibrio fischeri*: former *Vibrio fischeri*.

TABLE 2. Aquatic ecotoxicity tests performed with concrete and CDW leachate.

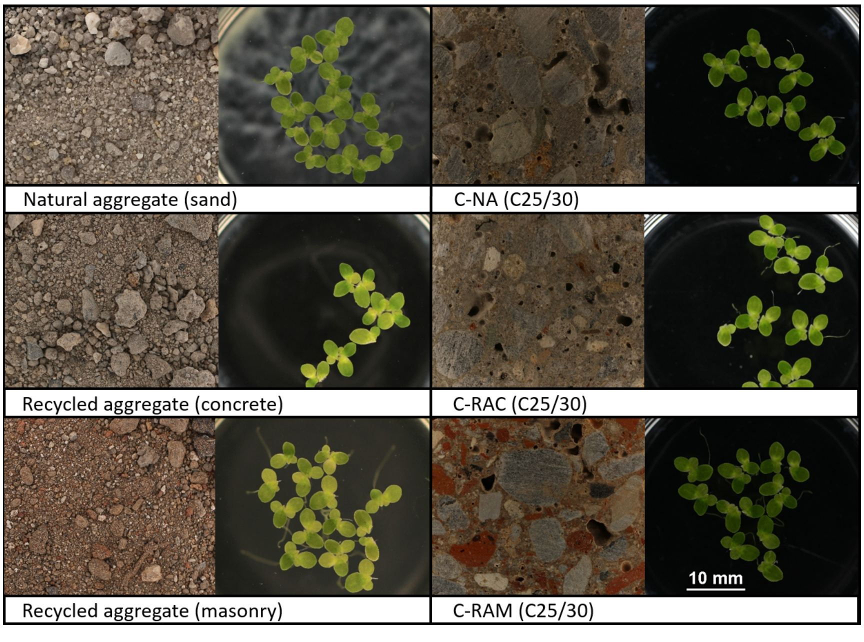


FIGURE 3. Example of *Lemna minor* semichronic toxicity test performed with 80 % leachates of natural and recycled aggregates fraction 0–4 mm (left) and 80 % leachates of concrete mixes (C25/30) based on the relevant aggregates (right). C-NA – reference concrete, C-RAC – concrete containing concrete-derived recycled aggregate, C-RAM – concrete containing masonry-derived recycled aggregate.

substances can be expected [14, 20]. Accordingly, for the selection or non-approval of certain types of concrete in aquatic environment, it is necessary to set stricter limits for concrete samples in the form of monoliths than for CDW or other homogenized materials. Hence, more studies on monolith samples are needed to compare the leaching potential of homogenized and uncrushed samples.

## 4. Conclusions

With the growing concern about the impact of construction products and recycled materials on the environment, interdisciplinary methods are needed. The impact on the environment can also be understood in terms of ecotoxicological potential. As hazardous waste materials cannot be disposed of on the landfill surface due to their high leaching potential, a more suitable variant seems to be their solidification in concrete. The use of CDW in recycled concrete must meet safety requirements. There are several standardized leaching protocols for construction materials and products. The application of aquatic biotests can be combined with the chemical analysis of leachates. However, the number of ecotoxicological studies is still very low. Further research is necessary to find effective methods that can be used in common practice.

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