UPSCALING THE POLLUTANT EMISSION FROM MIXED RECYCLED AGGREGATES UNDER COMPACTION FOR CIVIL APPLICATIONS

Adela P. Galvín^{1*}, Auxi Barbudo¹, Jesús Ayuso¹ and Manuel Cabrera

¹Department of Civil Engineering, University of Córdoba, Córdoba, Andalucía, 14001, Spain

Presenting author email: apgalvin@uco.es Phone and fax numbers: +34957212168; +34957218550

Abstract

In general terms, plant managers of construction wastes assess the materials according to concise leaching tests legally recommended that do not consider the compaction stage of the materials when they are applied on-site. Thus, the consequences of do not consider the compacted condition of recycled aggregates used at civil works (e.g. roads or embankments) leads to errors in the estimation of the pollutant potential of the material. For that reason, the present research designs an experimental procedure of leaching test for compacted materials (UCLT 2014). The aim of that laboratory test (designed specifically for granular materials used in civil engineering infrastructures) is evaluate the release of pollutant elements when the granulometry of the tested material is not modified and when the material is compacted at real work values. Apart from that laboratory test, the conventional standards: compliance leaching test (UNE EN 2457-3: 2003) and the Dutch percolation test (NEN 7343: 2004) were performed. After analysing the results, Chromium and Sulphate were released by the recycled mixed aggregates in higher amount, being considered as more conflictive pollutant elements. The release results of the designed leaching test were compared with the data obtained by conventional leaching test. In conclusion and based on the data, the release levels obtained from the UCLT 2014 were lower than the levels obtained with the conventional standards. It confirms that when the leaching behaviour is evaluated on construction materials without density alteration (an unreal situation for aggregates applied in engineering), the leaching behaviour is not accurately assessed.

Keywords: Sustainable construction, Leaching, Recycled Aggregates, Compaction.

Introduction

Application of recycled materials as a replacement for natural aggregates demands an assessment of the pollution potential of materials during its second life cycle. Through EU Landfill Directive, the European Commission has provided the foundations for the valorisation of such waste and established criteria to classify waste based on the potential contamination, as determined by leaching behaviour. When recycled aggregates are applied to civil works, they are exposed to rainwater, and potential pollutants may be leached and cause serious environmental problems.

Previous works focused on unbound applications have demonstrated that recycled materials possess suitable geotechnical properties for use in the construction of embankments and as granular sub-bases in road pavements [1, 2, 3].

The use of recycled aggregate from Construction and Demolition Waste (CDW) as alternative materials are limited an affected by the operations carried out by the treatment plants. Optimum management

operations result in a product (recycled aggregate) with optimum technical characteristics that also will not have negative environmental consequences in the environment after its application at works [4]

Thus, recycled aggregates could be predominately composed of inert components based on their origin (aggregates, concrete, wood, etc.). However, hazardous components, such as adhesives, paints, sealants, and equipment with PCBs, may be in contact with the material before it is separated from the aggregates in the treatment plant.

Spain legislation is complex and as one of the Member States of the European Union, the policy for C&DW management is strict. Thus, about legislation on environmental assessment, the Royal Decree 105/2008 [5] specifies that the total leachability and pollutant content of the waste and the ecotoxicity of the leachate must be controlled to prevent risks to the quality of surface water or groundwater. Plant managers operating in the Autonomous Community of Andalusia use the Territorial Management Plan of Non-Hazardous Waste of Andalusia as reference for the period 2010-2019 [6]. The management of C&DW leachates in landfills and the authorised treatment operations are included within the waste management framework developed by the Waste Catalogue of Andalusia, according to Decree 73/2012 [7].

To satisfy all those legal regulations, the daily operation of CDW plants includes the environmental assessment of recycled aggregates. In that sense, plant managers use procedures that are simple, cheap and easy to perform, obtaining laboratory results that are comparable with the actual legal limits. However, despite the conventional test are quick and easy to perform, these tests are not representative of construction material conditions. Granular materials used in civil engineering infrastructures (e.g. roads or embankments) have altered their physical conditions due to the compaction stage needed to support traffic loads increasing the bearing capacity of the material [8].

The importance of verify the relation between laboratory-field leaching results have previously been analysed by several authors [9, 10]. In most cases, it is confirmed that results of laboratory tests cannot directly translate to field conditions being necessary developed translation factors between the lab and field conditions to extrapolate the data to longer time scales [11].

The present research present a laboratory test (named as UCLT 2014) that have been designed considering the real stage of aggregates used in civil infrastructures. For that reason, the designed procedure test the samples without pre-crushing or alteration of the grain size distribution. In addition, the material is tested under the real compaction degree at work (e.g. as road sub-base).

The pollutant elements evaluated in the present study (twelve heavy metals and three anions) are the indicated by the European Council Decision 2003/33/EC [12] which establishes the criteria and procedures that are used for the admission of waste in landfills and the classification of these materials as inert, non-hazardous or hazardous.

Previous studies report the leaching behaviour of different RA from CDW. Galvín et al. [13] revealed that the compounds that present a leachability close to the limits for inert waste according to the EU landfill directive are Ni, Cr, Sb, Zn, Cu, and sulphate. Butera et al. [14] concluded that Se, Cr, Sb and sulphate are the most critical components. However, GEAR Project [15] analysed RA from a wide number of treatment plants in Spain and detected only Cr and sulphate as more critical components. This founding was

corroborated later by I del Rey et al. [16] which confirmed that Cr and sulphate were the major elements in recycled mixed aggregates from CDW. Therefore, based on the described framework, research works on mixed recycled aggregates has confirmed the need for further study. For that reason, the present work is focused on the analysis of the liberation of legally regulated elements. Particularly, the present study will analyse sulphate and chromium levels, but not only using standard and conventional leaching tests, but also evaluating the release without altering the granulometry of the tested material and being previously compacted (simulating the situation of the actual material on site). The results obtained confirm the need to develop tests closer to the real situation of material (e.g. materials as sub-base of roads).

2. Materials

A mixed recycled aggregate, named as MRA was selected for developing the present study. For its characterisation, several laboratory tests were performed and the results are listed below:

2.1. Classification test for the constituents (UNE EN 933-11:2009)

This method identifies and estimates the relative proportions of constituent materials of coarse recycled aggregates. The results are shown in the **¡Error! No se encuentra el origen de la referencia**..

Table 1. Properties of characterization of MRA					
Parameters	MRA				
Proportions of constituent materials					
Cement and products from cement (M _C)	48,18%				
Non-bound natural aggregate (M _U)	16,36%				
Brick and ceramic particles (M _B)	32.80%				
Other materials (M_X)	2,57%				
Light particles (M _L)	0,09%				
Density and water absorption					
Bulk density 4-31.5 mm	2.065 g/cm^3				
Water absorption 4-31.5 mm	12.163 %				
Bulk density 0.063-4 mm	2.827 g/cm^3				
Water absorption 0.063-4 mm	3.642 %				
Compaction-related test					
Compaction maximum density	1.81 g/cm^3				
Compaction optimal humidity	13.2 %				

Due to the building typology of the Mediterranean zone, it is very frequent to find high proportions of ceramic particles (coming from tiles, bricks, etc.) in the wastes coming from demolitions [15] [16]. It is corroborated by the results in Table 1 and according to the basic characterisation data, the material studied is mainly composed of mixtures of ceramics, stone aggregates, and, mainly, concrete products (mortar).

2.2. Density and water absorption test (UNE EN 1097-6:2001)

The procedure specifies the methodology for determination of particle density and water absorption (see data in *Table 1*). The test was performed both in the fine fraction (0.063-4 mm) and the coarse fraction (4-31.5 mm). Based on the data, a high absorption is observed mainly due to the recycled nature of the aggregate, with a higher content of fracture faces.

2.3. Particle size distribution test (UNE EN 933-1:1998)

The granulometric analysis plotted in *Figure 1* shows that it is an aggregate of continuous granulometry, well graded, with a maximum size of 25 mm.



Figure 1. Particle size distribution of MRA

2.4. Compaction-related test (UNE-EN 103-501:1994)

The aim is to determine the properties related to the soil compaction. This information can be used as a basis for specifying requirements for soils compacted in the field, and the maximum densities of granular soil and the moisture condition values can be calculated.

Thus, both parameters are showed in *Table 1*: the maximum density shows a high value around 13%, possibly due to the high absorption mentioned in the previous section.

3. Experimental methods

The present research is focused on the study of the pollutant release of the pollutant elements listed by EU Landfill Directive (heavy metals and anions indicated). The experimental procedure has been developed evaluating the environmental assessment of a recycled mixed aggregate, according to three different leaching methodologies performed in three phases of study, as it can be observed in Fig. 2:



Figure 2. Phases of the experimental procedure.

Phase 1: The compliance batch test for classification of the hazardous level of the MRA. The concentration data are compared with the legal limit established by the Landfill Directive.

Phase 2: The percolation test for estimation of the leaching behaviour in the short, medium and long term of aggregates without density alteration. The cumulative release curves were obtained.

Phase 3: The under compaction leaching test was designed for analysing the pollutant potential of granular aggregates considering the real conditions of materials used in civil infrastructures: high density due to compaction. The results can be easily compared with data from the conventional tests.

3.1. Compliance test for leaching of granular waste materials and sludges (UNE-EN 12457-3:2003)

The compliance test is a procedure of basic characterization of materials. The procedure consists of a twostep batch leaching test resulting in two liquid/solid ratios (L/S). This method use a solution of 175 g of dry mass of material (particle size <4 mm) with deionised water. In the first step, the solution is shaken for $6 \pm$ 0.5 h with an L/S of 2 l/kg. In the second step, water is added to stablish an L/S of 10 l/kg, and the solution is then shaken for another 18 ± 0.5 h. In both stages, the samples are left to decant during 15 min, the eluate is filtered (0.45 µm membrane filters), a subsample of 40 ml of eluate is collected for testing, and the pH, conductivity and temperature are measured.

3.2. Determination of the leaching of inorganic components from granular materials with the column test (NEN 7343:2004)

The Dutch column leaching test simulate the leaching behaviour of a waste material in the short, medium and long term by relating the amount of release that is expressed as mg/kg leached to the liquid/solid ratio. The relationship of the pollutant release with the time-scale is obtained from the height of the application and the infiltration rate. The procedure consists of a seven-step column test resulting in seven L/S ratios (0.1, 0.2, 0.5, 1, 2, 5, and 10 L/kg). The column (inner diameter of 5 cm and length of 20 cm) is filled with the test material (particle size <4 mm) and the initial dry matter is calculated. The deionised water quantity for each step is calculated from the dry matter and the L/S relationship. In the first step, a peristaltic pump (flow rate of 18 ml/h) fills the column with deionised water until the material is saturated, the eluate passes through two filters (1.5-µm prefilter and a 0.45-µm filter) to prevent entrainment of fine particles, and a collections flask picks up the leachate corresponding to each L/S ratio. From each flask, a subsample of 40 ml of eluate is collected for testing, and the pH, conductivity and temperature are measured.

3.3. Determination of the leaching of inorganic components from granular materials under compaction

The under compaction leaching test (UCLT) was designed with the purpose of analysing materials that will form part of infrastructures and must have an adequate load capacity. The main objective is to compare the obtained results with the data from standardized and conventional tests. The procedure consists of filling a stainless steel rectangular tank with the material (with the real grain size of a coarse aggregate: 0-32 mm). The dimensions of the tank are: 20 cm thick, 20 cm high, 50 cm wide and 100 cm long. The steel tank can be tilted with different positions (1, 2, 3 and 4% slope) through the manipulation of four support feet. In addition, this will have a smaller compartment to collect the leachate after the percolation of the leachant thought the aggregate. The compartment is separated from the tank by a perforated gate with grooves (*Figure 3*).

The MRA tested has 15 cm of thickness. With the maximum density results obtained in the Modified Proctor test (see Table 1), the dry mass of the tested material is calculated (assuming it will be compacted at 90% of the maximum density). The initial moisture of the material (6.39%) and the mass of water to be added to the stainless-steel tank are determined to achieve the optimum compaction moisture determined in the Modified Proctor test. The granular material and water are introduced into a blade kneader and mixed for 10 minutes. Meanwhile, a geotextile material is placed in the bottom of the drawer previously moistened.



Figure 3. Stainless-steel rectangular tank

Approximately half of the mixed material is added and the material is compacted with the Kangoo vibrating hammer. The second layer is added with the remaining material and compacted with the procedure described above (*see Fig 4*). Once compacted, rainfall events were applied to reach the same L/S fractions as in the column test NEN 7343: 0.1, 0.2, 0.5, 1, 2 L/kg. For practical reasons, the last two steps at L/S 5 and 10 1 / kg were not estimated. However, the long-term tendency curve can be elaborated with the first five steps. After each rain episode, leachates were drained into polyethylene containers through the drain at the base of the drawer. After each L/S ratio, the leachates are collected and pH, conductivity and temperature data are measured.



Figure 4. Sample preparation on UCLT test

After each leaching test, once suspensions are filtered two subsamples were removed from each leachate for determination of heavy metal and anion concentrations. Thus, the leachates were analysed by inductively coupled plasma mass spectrometry (ICP-MS) using a PerkinElmer ELAN DRC-e spectrometer for quantifying the 12 heavy metals specified by the European Landfill Directive: Ni, Cr, Sb, Se, Mn, Hg, As, Pb, Cd, Cu, Ba and Zn. In addition, the sulphate, fluoride and chloride anion contents were obtained by ion chromatography according to the requirements of standard UNE-EN ISO 10304-1: 2009.

4. Results

4.1 Evaluation of pollutant potential of RMA

The pollutant behaviour was analysed by the compliance test and classified according to the legal limits imposed by the European Landfill Directive [12]. Measured concentrations in the leachate (mg/kg) can be observed in *Table 2* (the data in bold indicate that the value exceeds the limit for inert wastes).

L/S 2 L/S 10 L/S 2 L/S 10 Cr 0.2 0.5 0.2062 0.2620 Ni 0.2 0.4 0.0068 0.0326 Cu 0.9 2 0.0232 0.0210	
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Ni 0.2 0.4 0.0068 0.0326 Cu 0.0 2 0.0232 0.0210	
C_{11} 0.0 2 0.0222 0.0210	
Cu 0.9 2 0.0232 0.0319	
Zn 2 4 n.d. n.d.	
As 0.1 0.5 0.0029 0.0092	
Se 0.06 0.1 n.d. n.d.	
Mo 0.3 0.5 0.0669 0.0775	
Cd 0.03 0.04 n.d. n.d.	
Sb 0.02 0.06 0.0017 0.0050	
Ba 7 20 0.0777 0.3156	
Hg 0.003 0.01 n.d. n.d.	
Pb 0.2 0.5 n.d. n.d.	
Cl ⁻ 4 10 $< 2 \text{ mg/l} < 2 \text{ mg/l}$	
F ⁻ 550 800 52.35 63.78	
SO ₄ ⁼ 560 1000 2488 11200	

Table 2. Leachate concentrations (mg / kg) obtained by the compliance test UNE EN 12457-3.

Footnote: n.d. non-detected and below the detection limit

Thus, comparing the measured concentrations of the MRA with the European environmental criteria at L/S 2 and 10 L/kg (see Table 2), the tested material is classified as non-hazardous waste due to the limit of inert wastes is exceeded for Chromium and Sulphate. The high presence of sulphate in the leachate is probably due to the high content of gypsum and ceramics present in the mixed aggregates [4] [16] [17]. Regarding the metal chromium, it has already been detected at high levels in other mixed recycled aggregates of similar characteristics, demonstrating in previous studies that the ceramic particles are the origin of this element in the leachates [13] [15].

4.1. Evaluation of pollutant release at the long term

After the basic characterisation of the release of contaminants by a compliance test and the identification of the most conflictive elements, the percolation test according to the Dutch procedure NEN 7343: 2003 was performed to evaluate the release of components under equilibrium or into leachate over time [18]. Compared with the compliance test, this leaching procedure is a closer approach to the processes occurring under field conditions. However, the main purpose is the characterization of the release but not the extrapolation of the leaching behaviour on a real situation [19] [11].

Table 3 shows the cumulative release of the regulated list of heavy metals and anions from an L/S of 0.1 to 10 L/kg and the regulated limits of the Landfill Directive for the first extraction of the column test at an L/S of 0.1 L/kg. The data in bold indicate that the value exceeds the limit for inert wastes

	Inert limit	L/S ratios (L/kg)						
	at 0.1 L/kg	0.1	0.2	0.5	1	2	5	10
As	0.006	0.0002	0.0007	0.0010	0.0017	0.0023	0.0038	0.0058
Ba	0.400	0.0041	0.0122	0.0317	0.0685	0.1154	0.2651	0.4808
Cr	0.010	0.0254	0.0522	0.0696	0.1176	0.1615	0.2255	0.2661
Cu	0.060	0.0028	0.0063	0.0085	0.0148	0.0202	0.0262	0.0283
Mo	0.020	0.0109	0.0237	0.0335	0.0519	0.0733	0.1017	0.1312
Ni	0.012	0.0007	0.0015	0.0021	0.0055	0.0060	0.0060	0.0060
Sb	0.010	n.d.	n.d.	0.0004	0.0009	0.0023	0.0057	0.0148
F-	0.250	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l
Cl-	46	22.66	43.5400	49.27	67.80	92.59	117.585	167.585
SO4=	150	160.62	481.300	1251.60	2752.8	4114.6	8450.25	15089.25

Table 3. Cumulative release (mg/kg) obtained according the percolation test NEN 7343:1995

Footnote: n.d. non-detected and below the detection limit

As it can be observed, in most of cases the released levels increase as the L/S ratio increases because the proportion of leachant and the contact time increases as the test is running. It is reconfirmed the higher levels released are those of sulfate. It may be due to the high content of gypsum and ceramic particles present in the sample (see Table 1). Again, the material is classified as non-hazardous waste based on the legal limits establish for L/S ratio of 0.1.

4.2. Evaluation of pollutant release under compaction

The cumulative release performing the designed test UCLT, are listed in Table 4.

	Inert limit	L/S ratios (L/kg)					
	at 0.1 L/kg	0.1	0.2	0.5	1	2	
As	0.006	0.0002	0.0005	0.0008	0.0016	0.0041	
Ba	0.400	0.0020	0.0058	0.0102	0.0188	0.0365	
Cr	0.010	0.0009	0.0026	0.0053	0.0089	0.0140	
Cu	0.060	n.d.	0.0003	0.0006	0.0006	0.0007	
Mo	0.020	0.0008	0.0019	0.0037	0.0058	0.0081	
Ni	0.012	n.d.	0.0003	0.0008	0.0015	0.0039	
Sb	0.010	n.d.	n.d.	0.0001	0.0002	0.0003	
F-	0.250	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l	<2 mg/l	
Cl-	46	0.57	1.40	2.82	4.85	8.85	
SO4=	150	33.90	82.7000	143.70	234.93	372.33	

Table 4. Cumulative release (mg/kg) obtained according the UCPT

Footnote: n.d. non-detected and below the detection limit

As it can be observed in Table 4, when the recycled aggregate is tested as follows: (1) with a real grain size (0-32 mm is a normal grain distribution for coarse aggregates used in roads) and (2) compacted at a value similar to the compaction degree in civil works (see values in Table 1), the release levels are lower than the registered with the conventional leaching tests. Therefore, under the effects of compaction, the release

levels of all elements are lower than the inert limit values. Thus, the RMA is in this case, classified as inert waste, and Cr and Sulphate are not released in hazardous levels. Then, it is necessary to graphically compare the cumulative release curve obtained by the conventional method (NEN 7343) and the curve obtained with the new designed leaching method (UCLT). The comparison of both release curves can be seen in Fig. 5.



Figure 5. Cumulative release curves of Cr and sulphates. Comparison between leaching tests..

Analyzing the cases of chromium and sulphate as the most conflicting elements in mixed recycled aggregates, it is verified that after compacting the material and testing it with a real granulometry, the release levels decrease. Therefore, in both cases, the cumulative release curves described by the proposed test UCLT are below the release curves obtained by the conventional percolation leaching method.

5. Conclusions

- Chromium and sulphate are identified as more conflictive elements since, based on compliance test data, they exceed the limit values established for inert waste. Therefore, the recycled mixed aggregate is classified as Non-Hazardous material according to the Landfill Directive.

- When long-term evolution of the pollutant release is analysed by means of a conventional test (NEN 7343) it is again verified that chromium and sulfate exceed the legal limit values for inert residues, and the RMA cannot be classified as Inert construction product.

-After proposing a leaching test in which the material is minimally altered in its granulometry and in which the RMA is compacted to work levels, great differences are detected with respect to the conventional test. Thus, despite applying the same L/S ratios as the standard percolation test, the registered cumulative release values were quite different. After comparing the long-term evolution of the release obtained with the NEN and the UCLT, it is verified that when the material is tested under a compaction state and at a normal granulometry of a coarse aggregate (0-32 mm), the release decreases and under these test conditions the RMA is classified as Inert material. This finding confirms the following: to evaluate the contaminant potential of granular materials used as structural component of civil infrastructures, they must be tested in laboratory under a compaction state. Therefore, it is necessary to evaluate them under near-real physical and mechanical conditions, being compaction degree directly related to properties such as: density, porosity, etc. [20] e.g. physical factors that undoubtedly affect the release of elements, as has been proved with the results obtained by the UCLT test.

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