

Article

Chemical and Leaching Behavior of Construction and Demolition Wastes and Recycled Aggregates

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Abstract: Construction and demolition wastes are widely recognized as the main waste stream in the EU, and their recycling and recovery is an important issue in sustainable building industry development. The composition of construction and demolition wastes is highly heterogeneous and is influenced by several factors, including the raw materials and construction products used. The environmental performance of these materials are therefore considerably variable and, in some cases, do not comply with the regulatory limits established to ensure the protection of the natural environment. In this context, this paper presents a data analysis on the environmental behavior of construction and demolition wastes and recycled aggregates in terms of both chemical composition and the release of contaminants according to a leaching test. Subsequently, the most critical parameters for recovery were identified and statistically evaluated. The leaching results showed that SO₄, Cu, and COD are critical compounds for both CDWs and RAs.

Keywords: construction and demolition wastes; recycled aggregates; chemical composition; leaching test

1. Introduction

Waste management is an environmental and social problem with marked social and technical interests, since its revaluation transforms it into recycled material, generating a new product feasible for use in a second life cycle. Economic activities are required in general greater efforts to reduce and prevent waste generation, contributing to the achievement of European Commission policies, such as the Circular Economy Action Plan implemented in 2019 towards a circular economy.

In the past, construction and demolition wastes (CDWs) were considered discarded and were disposed of in landfills. The current trends in waste management systems to replace removal with valorization reflect the potential of waste as a resource rather than a problem [1]. In that sense, recycled aggregates from CDWs, which are mainly composed of concrete, natural aggregates, bricks, and to a lesser extent other constituents, such as gypsum, wood, glass, and plastics, have demonstrated their technical feasibility in construction works, such as bases and subbases of roads and backfilling, and other works, such as mortars, concrete, and beds of pipes or green applications [2–5].

CDWs represent one of the largest waste streams in the European Union by weight and volume. In 2018, the total waste generated by all economic activities and households (EU-27) amounted to 2317 million tons; this was the highest amount recorded during the period of 2008–2014 [6]. According to Eurostat data, the waste from the construction sector accounts for around 37% of total waste

production, with a value of 972.6 million tons in 2018. Italy, with a production of about 60.5 million tons, is the fourth European country for CDW production after France, Germany, and the Netherlands (not considering the UK, recently released from the EU), representing about 34% of the total waste produced in the nation (i.e., the sum of waste produced by all economic activities and households) in the same year [7,8].

Regarding recovery, some countries, such as the Netherlands, Germany, Denmark, and Austria, have achieved recycling rates of over 85% [9]. In this context, according to the Directive 2008/98/EC (amended by Directive EU 2018/851), which requires a minimum of 70% (by weight) of non-hazardous CDW recovery rate by 2020, the average recovery rate of the EU (27) is 47% [10]. In Italy, 77% of CDWs were recycled at the national level in 2018 [7]; therefore, the target of 70% seems to be achievable by 2020. However, the amount of CDWs destined for landfill is still important (24%). At the same time, other sources report a lower recovery rate of about 10% [11]. This is because 77% refer to CDW streams treated and stored but not yet actually recovered and used in real applications. The treated materials are, in fact, generally accumulated in treatment plants without concrete outlets in the economic market. Large storage areas at treatment plants have essentially become temporary landfills [12].

This is mainly due to several obstacles to sustainable CDW management, identified as follows [13]: (1) lack of confidence of stakeholders in the use of products derived from waste due to the various origin of RAs; (2) lack of knowledge on the environmental and technical characteristics of recycled aggregates (RAs) reduces the use of recycled materials; (3) uncertainty about the potential health risk for workers using recycled materials; (4) RAs are generally not competitive as compared to natural aggregates and, finally, (5) lack of end of waste criteria for the evaluation of RA eco-compatibility.

Moreover, as stated by Gálvez-Martos et al. [12] and Ayuso et al. [14], in some member states, there is a significant amount of illegal dumping, which hinders the development of the recycled materials market, that may not be reflected in official statistics.

CDWs arise from construction and total or partial demolition activities, and are codified by Chapter 17 of the European List of Waste. After a mechanical treatment process carried out through technologically interconnected phases of crushing, separation of metal and undesired fractions, screening, and particle size selection, new materials defined as recycled aggregates are generated.

The composition of CDWs is extremely variable due to the current demolition techniques, which can reduce the time and cost of the process compromising the debris homogeneity, the heterogeneity of building structures, and the lack of adequate treatments for RA production (CDWs are mainly treated in dedicated mobile plants that carry out rough treatments that lead to low quality RAs). However, there is a high potential for recycling and reuse of CDWs, since some of their components have a high resource value. Currently, in the construction sector, the RAs produced are mainly utilized in low-grade applications, such as road construction, pavement, and drainage [15]. In Italy, the national legislation for waste recovery is defined in the Legislative Decree 152/2005, amended by the Legislative Decree 205/2010. The specific environmental regulation governing the recovery and reuse of special waste, such as CDWs, is the Ministerial Decree (M.D.) 05/02/1998, subsequently amended by the Ministerial Decree 186/2006. In order to guarantee environment protection, this ministerial decree establishes specific limit values on the release of contaminants from recycled materials directly reintroduced into the environment in an unbound form (i.e., road and geotechnical applications). In particular, the materials must be subjected to a leaching test according to EN 12457-2 ($d \leq 4$ mm; $L/S = 10$; 24 h; demineralized water). In terms of environmental performance, the literature studies highlighted that chromium and sulfate are the most critical compounds in CDW and RA leachates [16,17]. Specifically, total chromium is mainly released by ceramic materials and partly carbonated samples [18], while high sulfate levels are released by gypsum-based materials [19] and other CDW compounds, such as mortar particles [20]. High concentrations of chlorides are sometimes recorded [5,16]. The high release is due both to the presence of cement and to release from ceramic materials. For the use of RAs for concrete production, chlorides must be kept under control, since over time they can lead to corrosion of the reinforcing bars [21]. In addition, several studies highlighted that the release

of contaminants is also influenced by other factors that regulate the leaching process, such as the pH and carbonation grade, liquid to solid ratio, particle size, or contact time [22–26]. The leaching behavior in construction materials is mainly controlled by the alkaline nature and acid buffering capacity of the stabilized matrix, and metal release presents a pH-dependent process controlled by metal hydroxide solubility [25]. The pH and the carbonation degree are, therefore, key elements to be analyzed, as they strongly influence the release of contaminants. Despite the high influence of pH dependency on leaching, the remaining factors mentioned above must be considered. In particular, the particle size of the examined solid determines the surface exposed to the leaching solution, which in many cases influences the release of the contaminant into the solution. In fact, a reduction in particle size leads to an increase in the contact area, which results in a greater release of pollutants [22,23,27]. Mahedi et al. [28] confirmed the previous studies, but also stated that the RCA particle sizes had a significant influence not only on the leached concentrations of elements, but also on the effluent pH and alkalinity. As stated above, the liquid to solid ratio (L/S) also represents a strong influence parameter. In particular, increasing the L/S increases the release of metals, according to Galvin et al. [29]. The same results were obtained by other authors [23,30] who evaluated the leaching release of As, Ba, Cu, Mo, Ni, Pb, Sb, Se, and Zn at an L/S equal to 2 and 10 L/kg. The results showed that all of the compounds analyzed had higher concentrations at a higher L/S ratio from 0 to 60 L/kg but, when the L/S ratio exceeded 60 L/kg, an equilibrium was reached and concentrations in extractants remained constant.

Finally, as stated above, the contact time may influence the amount of leaching contaminant. According to Lopez Meza et al. [31], shorter contact time has no significant impact on pH, conductivity, and release of contaminants in a long-term prediction. In contrast, Galvin et al. [29] highlighted that the longer stirring time causes a greater release of contaminants due to an increase of the contact time between leachant and material. The results showed that the highest level of contamination was obtained experimentally by the Dutch procedure NEN 7341:1994 after 6 h with reference to the one done after 3 h of stirring; the reason is the aggressive conditions imposed by this procedure in order to obtain the amount that is available for leaching (availability) in the tested material.

In this context, in the literature, there are many experimental studies carried out on CDWs and RAs aimed at evaluating the environmental performance of these materials [18,19,23,32–36], while there are very few studies that evaluate these properties using a statistical approach [37]. Therefore, the objective of this study is to evaluate the influence of the following parameters on the leaching behavior of CDWs and RAs by state of the art analysis and, simultaneously, assess the environmental behavior of both CDWs and RAs by analyzing characterization tests (e.g., chemical composition and leaching test) carried out on such materials. Special attention was given to the leaching properties in order to identify the critical parameters for recovery under the Italian national legislation.

2. Materials and Methods

2.1. Construction and Demolition Wastes (CDWs) and Recycled Aggregates (RAs)

CDWs consist of debris generated during construction, renovation, and demolition of buildings, roads, and other civil engineering structures. They are a large source of secondary raw materials, consisting roughly of concrete, wood, masonry, drywall, glass, plastics, metals, and more [38]. The recycling of this waste stream generally takes place at mobile or fixed treatment plants from which RAs are produced. Usually, mobile plants do not apply technologically advanced treatments (only metal separation and a volume reduction process), while fixed treatment plants are equipped with more performing technologies than mobile plants. The quality of the produced RA directly depends on the purity of the original CDW. Depending on the merceological fractions contained, the CDWs are classified with different EWC codes of chapter 17. Generally, in Italy, during the treatment process, all managed EWC codes are usually mixed together to produce mixed recycled aggregates (MRAs) of medium to low quality, mostly used in road and geotechnical applications.

In the present study, the CDWs most managed by the Italian treatment plants and the RAs produced were analyzed in terms of chemical composition and leaching behavior. The EWC codes analyzed are reported in Table 1.

Table 1. EWC codes of the CDWs analyzed.

EWC	Description
170101	Concrete
170504	Soil and stones other than those mentioned in 170503
170904	Mixed construction and demolition wastes other than those mentioned in 170901, 170902, and 17003

2.2. Chemical Composition and Leaching Behavior Data

In order to evaluate the environmental behavior of both CDWs and RAs, several certificates of chemical characterization and leaching tests were provided by the Italian National Builders' Association (ANCE Lombardy) and the National Association of Recycled Aggregates Producers (ANPAR). These certificates are related to CDWs and RAs treated by different treatment plants located in Italy, and give information about the chemical and leaching properties of CDWs and RAs according to the Italian environmental legislation (L.D. 152/2006 and M.D. 186/2006).

A total of more than 50 certificates were collected, of which 15 were on chemical analysis and 39 were on leaching tests (Table 2). Elemental composition of major and minor elements was determined according to the standards CNR IRSA 1 Q64 Vol. 3 1985 (pH), CNR IRSA 2 Q64 Vol. 2 1984 (dry residue at 105 °C), and EPA 3051A:2007+EPA6010C:2007 (metals). In particular, for metals analysis, the laboratory samples were prepared according to EPA 3051A:2007, which provides a rapid microwave acid extraction to mimic extraction using conventional heating with nitric acid (HNO₃); while the metal determination was carried out by inductively coupled plasma atomic emission spectrometry (ICP-AES), according to EPA6010C.

Table 2. Number of chemical and leaching test certificates in relation to the materials analyzed.

Analysis	Sample	
	CDWs	RAs
Chemical composition	14	1
Leaching test	27	12

Regarding the leaching behavior, the tests were performed according to the UNI EN 12457-2 (Figure 1). The procedure described by this standard consists of a one stage batch leaching test at a liquid to solid (L/S) ratio of 10 L/kg for materials with a particle size below 4 mm. The solution, composed of granular material and deionized water, is shaken for 24 ± 0.5 h and then left to decant for 15 minutes. After that, the eluate is filtered using a membrane filter (0.45 µm), and a subsample is collected for testing. Conductivity, pH, and temperature must be immediately measured. The contaminant releases were then compared to the regulatory limit values imposed by M.D. 186/2006.



Figure 1. Tested samples and eluates produced.

2.3. Data Analysis

The chemical and leaching properties have been discussed in tabular format (range of maximum and minimum values) in order to identify the variation of the pollutant concentration. A more in-depth analysis was developed on data related to leaching tests, which were analyzed and statistically elaborated using the software IBM SPSS. In particular, the statistical analysis was developed by applying the box plot methodology in order to visually reproduce the variation of pollutant concentration and, at the same time, to identify the criticality level of the contaminants with respect to the regulatory limits for recovery. Box plots are useful in research methodology and are interesting, as they pictorially convey a large amount of information in a concise way that allows for quick data interpretation and understanding [39]. The typical construction of the box plot divides the data distribution into quartiles, as shown in Figure 2.

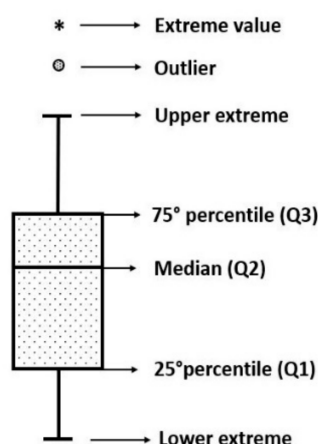


Figure 2. Box plot scheme.

A box is used to define the positions of the first (Q1) and third (Q3) quartiles, and the inside of this box indicates the interquartile range (IQR), which is the area between the first and third quartiles and contains 50% of the data population. The box is also intersected by a horizontal line, which represents the second quartile (Q2) and is defined as the median. Vertical lines, called whiskers, are extended to the extremes of the distribution, and represent the minimum and maximum values in the dataset.

The point and star represent, respectively, the outliers and the extreme values. These values are observations that lie an abnormal distance from the other population values.

3. Results and Discussion

3.1. Chemical Composition of CDWs and RAs

The minimum and maximum total content of chemical parameters of both CDWs and RAs are summarized in Table 3. Referring to CDWs, Al, Fe, Cr, Mn, and Si were the major most abundant elements, with maximum contents of 12,021 mg Al/kg, 211.30 mg Fe/kg, 277.70 mg Cr/kg, 1070 mg Mn/kg, and 421,000 mg Si/kg, respectively. Pb and Zn were also present in considerable amounts, with maximum concentrations of about 236 mg Pb/kg and 800 mg Zn/kg, respectively.

Sb, As, Co, Ni, Cu, Ba, Se, and V were present in minor concentrations (< 90 mg/kg), while other elements, such as Be, Cd, Cr (VI), Me, Mo, Sn, benzene, and asbestos were below the limit of detection in all of the analyzed data.

Referring to RAs, the most abundant major elements were Al, Cu, Cr, and Zn, with a maximum value of concentrations of 9442 mg Al/kg, 67.60 mg Cu/kg, 55.40 mg Cr/kg, and 50.30 mg Zn/kg, respectively.

All of the other elements were present, but in low concentrations, while Me, Fe, Mn, Mo, Ba, Si, Se, and benzene were not detected (n.d.).

Table 3. Total content of major and minor elements in CDWs and RAs (pollutant content reported in the minimum–maximum range).

Element	U.M.	CDWs		RAs
		Min	Max	
Dry residue at 105°	%	83.9	100	83.10
pH	-	6.80	11.22	n.d.
Aluminum	[mg/kg]	502.30	12,021	9442
Antimony	[mg/kg]	5	10.8	<10
Arsenic	[mg/kg]	1	27.8	11
Beryllium	[mg/kg]	1	<14	<10
Cadmium	[mg/kg]	1	<14	<10
Cobalt	[mg/kg]	0.10	14	<10
Total chromium	[mg/kg]	4.80	277.7	55.40
Chromium (VI)	[mg/kg]	0.10	<50	<10
Iron	[mg/kg]	211.30	211.30	n.d.
Mercury	[mg/kg]	0.10	<14	<10
Nickel	[mg/kg]	1	86.9	14.20
Manganese	[mg/kg]	0.10	1070	n.d.
Lead	[mg/kg]	0.50	236	33.45
Molybdenum	[mg/kg]	0.10	<14	n.d.
Total Copper	[mg/kg]	1	153	67.60
Barium	[mg/kg]	1	167	n.d.
Tin	[mg/kg]	0.10	<14	<10
Silicon	[mg/kg]	240	421,000	n.d.
Vanadium	[mg/kg]	12	80	<10
Selenium	[mg/kg]	0.10	13.3	n.d.
Zinc	[mg/kg]	5	800	50.30
Mineral Oils	[mg/kg]	0.50	5800	11.60
Benzene	[mg/kg]	0.10	<10	n.d.
Asbestos	[mg/kg]	<1000	<1000	<1000

3.2. Leaching Behavior of CDWs and RAs

As previously mentioned, the leaching behavior was evaluated according to the UNI EN 12457-2, and the results were compared to the legal limits imposed by M.D. 186/2006 for unbound recovery. Table 4 shows the leaching concentrations of both CDWs and RAs in terms of minimum and maximum values; while Figure 3, Figure 4, Figure 5, and Figure 6 represent the box plots of the main critical parameters for recovery (sulphates, total chromium, and COD). All parameters with maximum concentrations above the regulatory limits are marked in bold.

Table 4. Leaching concentration of CDWs and RAs (pollutant content reported in the minimum–maximum range).

Element	U.M.	CDWs		RAs		Limit (M.D.186/2006)
		Min	Max	Min	Max	
pH	-	6.60	11.30	7	11.50	5.50–12
Nitrates	[mg/L]	0.05	27	1.20	29.10	50
Fluoride	[mg/L]	0.08	2.09	0.13	0.50	1.50
Sulphates	[mg/L]	12.40	1613	13.90	615	250
Chloride	[mg/L]	0.77	186	3.10	16.60	100
Cyanide	[µg/L]	5	20	5	20	50
Barium	[mg/L]	0.002	0.34	0.01	0.10	1
Copper	[mg/L]	0.001	0.067	0.002	0.08	0.05
Zinc	[mg/L]	0.005	0.058	0.001	0.01	3
Beryllium	[µg/L]	1	5	0.10	5	10
Cobalt	[µg/L]	1	10	0.40	5	250

Table 4. Cont.

Element	U.M.	CDWs		RAs		Limit (M.D.186/2006)
		Min	Max	Min	Max	
Nickel	[µg/L]	1	14.90	1	6.20	10
Vanadium	[µg/L]	0.15	199	0.015	34	250
Arsenic	[µg/L]	1	11	1	5	50
Cadmium	[µg/L]	0.20	4.80	0.10	1	5
Total Chromium	[µg/L]	1	94	5	42	50
Lead	[µg/L]	1	78	0.10	5	50
Selenium	[µg/L]	1	5	0.60	7	10
Mercury	[µg/L]	0.1	1	0.10	0.50	1
Asbestos	[mg/L]	1	10	1	5	30
COD	[mg/L]	2	93.70	5	40	30

Numbers marked in bold indicate parameters with maximum concentrations above the regulatory limits.

The material pH and the pH of its environment are crucial in determining the release of many constituents. The results highlighted that pH is generally alkaline for both CDWs and RAs. As shown in Figure 3, CDWs from concrete (EWC 170101) are the main CDWs responsible for the high alkalinity due to cement, as stated by Del Rey et al. [18]. However, while remaining alkaline, the pH may change in relation to the characteristics and age of the concrete contained in the analyzed material. As stated by Braga Maia et al. [24], RAs from young concrete have strongly alkaline pH values ($\text{pH} > 13$), while RAs generated from older carbonated concretes have lower pH values ($\text{pH} < 10$). In fact, pH is strictly related to the carbonation degree, which is linked to the age of the building, the exposure conditions during its lifetime, and the type of concrete [28]. In particular, carbonation occurs when the cement hydrate phases react with CO_2 , precipitate as calcium carbonate (CaCO_3) [40], and develop carbonated material [41]. This process generally proceeds slowly during the service life of concrete structures, but when these concrete materials are demolished and transformed into recycled aggregates, their specific surface increases, resulting in greater CO_2 absorption. In this context, Engelsen [42] stated that aging and carbonation processes change the material pH from $\text{pH} = 12\text{--}14$ to $\text{pH} = 9\text{--}10$, which significantly modifies the leachability of many chemical substances. Butera et al. [16] demonstrated that this decrease of pH, due to the carbonation, increases SO_4 , Si, Cr, and V releases and decreases Ca, Ba, Sr, Na, and K releases. The results of the study also showed that most of the total Cr was released as Cr (VI), which is more harmful and leachable than Cr (III) when the pH is less than 5 [18,43].

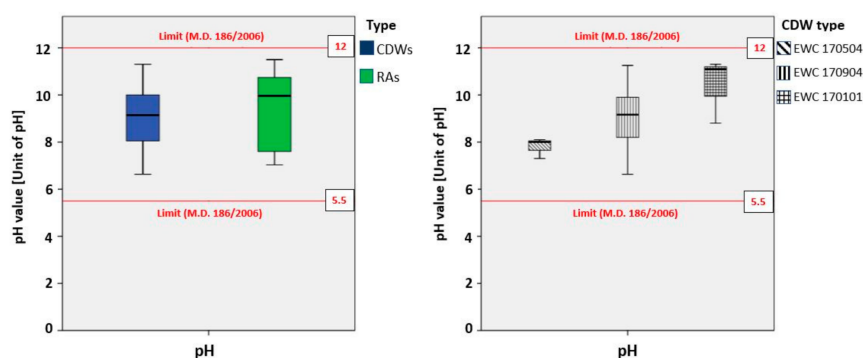


Figure 3. pH values: comparison between CDWs and RAs (left) and comparison among different CDWs (right).

We identified the most conflictive elements; Figures 4–6 show concentrations (expressed in mg/L) of SO_4 , COD, and Cr released in high levels by both material CDW and RA.

As shown in Figure 4, SO_4 is a critical parameter for both CDWs and RAs, and it is generally released from mixed CDWs due to the presence of gypsum, ceramic materials, and mortar

particles [17,19,20,23,30,36]. In particular, as shown in Figure 4 (right), over 50% of all samples analyzed (box with vertical lines) had concentrations above the regulatory limit of 250 mg/L. Moreover, some concentrations, classified as outliers, reached values over 1400 mg/L and 1600 mg/L. The correlation between SO_4 and gypsum was also proved by Lopez-Uceda et al. [35], who found that samples with high gypsum content do not meet regulatory requirements due to the high sulfate concentration. In this context, even though gypsum is a limiting element in RAs, this research has supported its long-term feasibility in terms of physical, mechanical, and leaching properties when used in road construction.

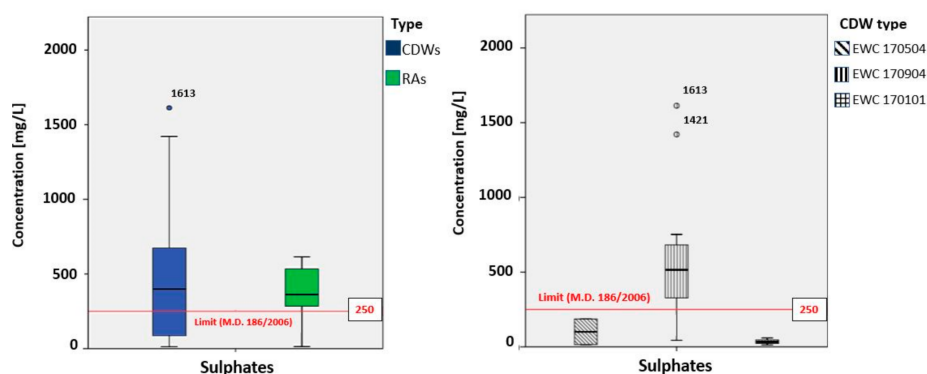


Figure 4. SO_4 concentrations: comparison between CDWs and RAs (left) and comparison among different CDWs (right).

As shown in Figure 5, Cr is mainly released by cement and concrete fractions (EWC 170101), confirming the results obtained by Chen et al. [33], Vegas et al. [10], and Nurhanim et al. [44]. However, these results are not in line with what was stated by Del Rey et al. [18], who highlighted that the release of Cr and SO_4 is not correlated to concrete samples. Similarly, Galvin et al. [23] stated that Cr concentrations are correlated with ceramic particles (mainly from bricks and tiles).

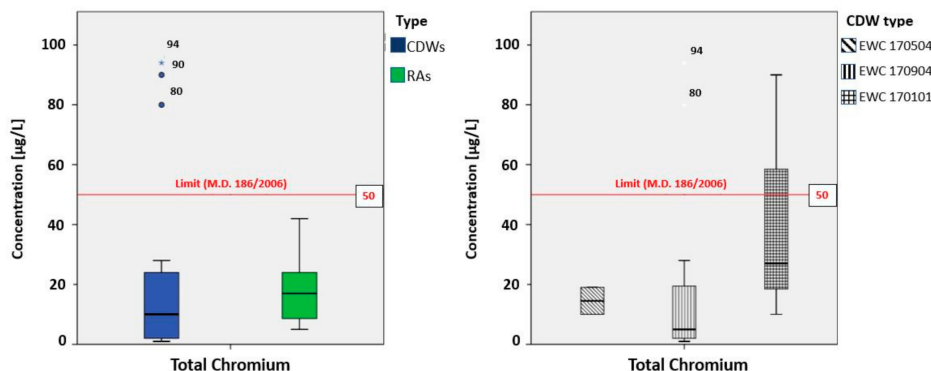


Figure 5. Total Cr concentrations: comparison between CDWs and RAs (left) and comparison among different CDWs (right).

Finally, COD, as one of the most critical parameters, was released by both cement and concrete CDWs (EWC 170101), soil and stones (EWC 170504), and mixed CDWs (EWC 170904). In particular, among them, soil, stones, and concrete are the main sources of release, with median values of about 45 mg/L and 58 mg/L, respectively. In fact, as shown in Figure 6, all soil samples exceeded the legal limit value of 30 mg/L, as the whole box and the whiskers reside above the limit. As stated by Melendez [45], these high levels of COD are attributed to the decomposable organic matter that could be present in both CDWs and RAs.

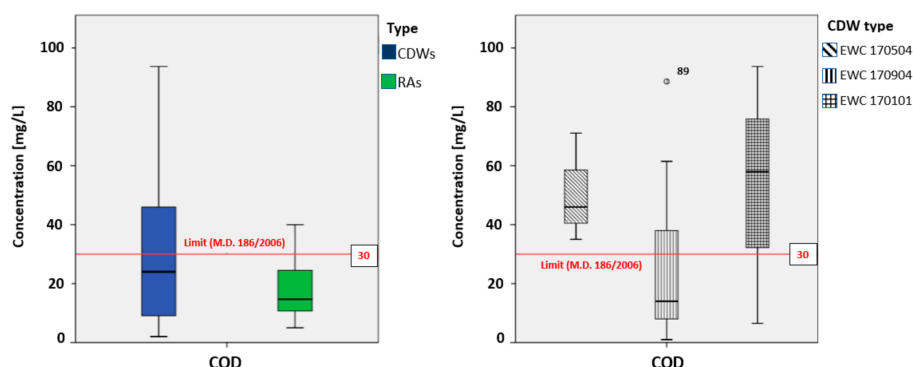


Figure 6. COD concentrations: comparison between CDWs and RAs (left) and comparison among different CDWs (right).

4. Conclusions

In this study, the environmental performances of CDWs and RAs in terms of chemical composition and leaching behavior were evaluated. The chemical results showed that Al, Fe, Cr, Mn, and Si are the most abundant parameters contained in CDWs, while Al, Cu, Cr, and Zn are the most abundant parameters contained in RAs.

Comparing the leaching results with the regulatory limit values of M.D 186/2006 led to the following conclusions:

- F, Cl, Ni, Cr, and Pb are critical parameters only for CDWs;
- SO_4 , Cu, and COD are critical parameters for both CDWs and RAs;
- SO_4 is released mainly from mixed CDWs (EWC 170904) due to the presence of gypsum, ceramic materials, and mortar particles;
- Cr is released by concrete fractions (EWC 170101) due to the presence of cement;
- COD is released by cement and concrete CDWs (EWC 170101), soil and stones (EWC 170504), and mixed CDWs (EWC 170904).

In conclusion, the critical parameters that affect the potential recovery and reuse of both CDWs and RAs are still numerous. As revealed by the analysis performed, the release of these compounds is governed by the merceological fractions contained in the materials analyzed. Therefore, in order to improve the environmental quality of CDWs and, consequently, of RAs, the application of a selective demolition process is of primary importance.

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