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Photocatalytic Recycled Mortars: Circular Economy as a Solution for Decontamination

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Abstract: The circular economy is an economic model of production and consumption that involves reusing, repairing, refurbishing, and recycling materials after their service life. The use of waste as secondary raw materials is one of the actions to establish this model. Construction and demolition waste (CDW) constitute one of the most important waste streams in Europe due to its high production rate per capita. Aggregates from these recycling operations are usually used in products with low mechanical requirements in the construction sector. In addition, the incorporation of photocatalytic materials in construction has emerged as a promising technology to develop products with special properties such as air decontamination. This research aims to study the decontaminating behavior of mortars manufactured with the maximum amount of mixed recycled sand without affecting their mechanical properties or durability. For this, two families of mortars were produced, one consisting of traditional Portland cement and the other of photocatalytic cement, each with four replacement rates of natural sand by mixed recycled sand from CDW. Mechanical and durability properties, as well as decontaminating capacity, were evaluated for these mortars. The results show adequate mechanical behavior, despite the incorporation of mixed recycled sand, and improved decontaminating capacity by means of NO_x reduction capacity.

Keywords: recycled aggregates; recycled mortar; construction and demolition waste; decontaminating; photocatalysis

1. Introduction

A current environmental challenge is to optimize natural resources. For this, minimizing and recovering waste materials are essential from a productive point of view of the main economic sectors. The circular economy model is a sustainable production strategy where the reuse of waste as secondary raw materials is underlined, achieving comprehensive management of waste materials [1]. So, the circular economy concept describes a cyclical system in which economic and environmental aspects are integrated [2]. Reducing the resources used and the waste generated can save resources and help reduce environmental pollution.

At the European level, this model is implemented through the Action Plan for the Implementation of the Circular Economy of the European Parliamentary Commission [3]. This proposed transition toward a more circular economy brings great opportunities. It is important to make efforts to modernize and transform the economy, shifting it toward a more sustainable direction, which will enable companies to make substantial economic gains and become more competitive in the market. This not only offers important energy savings and environmental benefits, but also creates local jobs and opportunities for social integration. The wider benefits of the circular economy also include reduced energy consumption and carbon dioxide emission levels [3].

The model highlights the important role played by the construction sector, which is still associated with strong negative environmental effects due to the high natural resource consumption for manufacturing and the large production of waste [4]. Waste from construction activities, including excavation or land formation, earthworks, civil construction and building, road works, and building renovation, are considered as construction and demolition waste (CDW) [5,6]. These represent a relevant part of the total generation of solid waste worldwide [7]. Due to this, European Directive 2008/98/EC [8] established that EU member states must reach a recycling rate of 70% for CDW in 2020 to avoid sending it to landfills. However, the potential for reuse and recycling of this waste stream is not being fully exploited. One obstacle is a lack of confidence in the quality of recycled construction and demolition materials. For this reason, on 9 November 2016, the European Commission proposed an industry-wide voluntary protocol on the management of construction and demolition waste [3]. The aim of the protocol was to improve the identification, source separation, and collection of waste, as well as logistics, processing, and quality management. The protocol could thus increase trust in the quality of recycled materials and encourage their use in the construction sector.

After CDW is properly treated, its use as recycled aggregates (RAs) in the construction sector has been widely addressed to mitigate environmental problems such as the consumption of raw materials and waste landfilling. This leads to an increase the recycling rate [9–15], becoming a practical reality that has allowed the development of specific regulations [16].

RAs have different physical, mechanical, and chemical properties from natural aggregates (NAs). They have lower density, higher water absorption, lower resistance to fragmentation, and a higher content of sulfur compounds and soluble salts [17,18]. Depending on the original waste material, recycled aggregates could be concrete, ceramic, or a mixture (recycled mixed aggregate, RMA). RMAs come from building demolitions and contain a wide range of materials, such as concrete waste, pavement material, ceramic products, and, in smaller quantities, other materials such as gypsum, glass, wood, etc. [19]. On some occasions, they are used in works with lower requirements, such as roads with low traffic intensity [20,21], bike lanes [22], urban and pedestrian roads, and unpaved rural roads [23–25], where RAs provide similar functional and structural characteristics as NAs.

Another widely studied application is in the manufacture of concrete. There are studies that support its use, even for structural concrete [26,27]. The use of recycled aggregates in the production of concretes and mortars has the following competitive advantages: (i) decreased extraction of aggregates from rivers, coasts, and quarries; (ii) exclusion of aggregates from landfills, reducing the volume of waste to be treated; and (iii) implementation of the circular economy model and approach to set recycling targets [14].

In most of these studies, only the coarse fraction was used [7,11,12,28]. It was concluded that concrete strength decreased when recycled concrete was used and the reduction could be as low as 40% [29]. No decrease in strength was reported for concrete containing up to 20% fine or 30% coarse recycled aggregates, but beyond these levels, there was a systematic decrease in strength as the content of recycled aggregates increased [29].

Of the different types of RAs, recycled crushed concrete (RCA) is the most widely used in the manufacture of new concrete [9,26,27,30–32]. Even the Spanish concrete standard (EHE-08) [16] allows the use of up to 20% RCA, but only referring to fractions greater than 4 mm. Because of the lower density of RMA, concrete made with it has lower density and higher water absorption than reference concrete [19].

Regarding the influence of recycled coarse and fine fractions, studies on recycled concrete incorporating fine recycled aggregate (FRA) from CDW did not obtain satisfactory results [19,29,30,32,33]. Kathib [29] studied the incorporation of FRA, and showed that properties such as density, dynamic modulus of elasticity, and compressive strength were reduced, the latter resulting in a decrease of 10% and more than 15% with a 50% and 100% incorporation ratio, respectively, at

90 days, in agreement with the results by Kou et al. [33]. Because of these differences, the use of fine fractions in concrete should not be dismissed, but more research on it is needed.

For this reason, the main application of FRA is in recycled mortar, for which the requirements are more tolerant. Within this group, there are different uses depending on where it will be placed: masonry mortar, interior or exterior rendering mortar, or mortar for the manufacture of paving blocks. Most of the investigations carried out to date were aimed at incorporating FRA in mortar for masonry [31,34–36]. However, this research in particular is intended for the use of recycled mortar for any application in contact with the atmosphere, such as wall cladding or paving blocks. With the results obtained, it is hoped that its use can be further expanded.

As in the case of recycled concrete, the use of mixed FRA (MFRA) in mortar has been found to be harmful to the properties of mortar [14,36]. However, this research aims to prove its use as suitable if it meets a series of quality requirements, normally linked to correct CDW treatment.

In addition to the circular economy, the mortar manufactured in this research is intended to provide a second benefit for environmental sustainability: atmospheric decontamination and reduced carbon footprint. The atmospheric pollution produced by accelerated population growth and industrialization can cause serious damage to the health of both people and ecosystems, and even to infrastructures and historical heritage. Among the main harmful emissions are nitrogen oxide gases (NO and NO_2, commonly known as NO_x) generated by transportation and various industries. These gases have high toxicity and can cause serious problems in human health, as well as environmental problems (acid rain, photochemical smog, destruction of the ozone layer, etc.) [37].

It is essential to reach the highest air quality that does not create a risk to people's health and does not cause deterioration or permanent damage to ecosystems. The current measures for reducing air pollution in cities associated with CO_2 emissions and other pollutant gases fall within two lines of action: citizen awareness policies, which advocate avoiding the use of private vehicles in favor of alternative means of transport, such as bicycles or public transport; and policies restricting the circulation of these vehicles, either with speed reductions or by prohibiting movement in downtown areas, as is carried out in cities such as Paris, London, and Madrid [38].

However, these measures are not fully effective in eliminating or reducing air pollution in urban environments, which is why complementary measures are needed in addition to conventional pollution control methods. Within these new measures, it is interesting to consider the advantages of photocatalysis.

The photocatalysis process starts from the natural principle of decontamination of nature itself. It is a similar technology to that of photovoltaic solar panels [39]. Like photosynthesis, which, thanks to sunlight, can remove CO_2 to generate organic matter, photocatalysis removes other usual pollutants in the atmosphere such as NO_x and SO_x (inorganic compounds) and volatile organic compounds (VOCs) through an oxidation process activated by solar energy. Through photocatalysis, most of the pollutants present in urban areas can be reduced, such as NO_x, SO_x, VOCs, CO, methyl mercaptan, formaldehyde, chlorinated organic compounds, poly-aromatic compounds, etc., which are aggressive in terms of both the properties of the material and the environment.

Construction materials treated with photocatalysts reduce, above all, NO_x particles that are produced by vehicles, industry, and energy production. During photocatalysis, the photocatalyst agent absorbs light energy, transfers it to a reactive compound, and triggers a chemical reaction through the formation of radicals. Titanium dioxide (TiO_2) and the products derived from it are the most widely used photocatalysts, and it is these that trigger the transformation of NO_x (nitrogen oxides) into nitrate through the action of sunlight. This can then be used to increase the shelf life of cement-based materials, while it can also be used to substantially decrease the concentration of some air pollutants, especially in semi-enclosed places such as important urban avenues, tunnels, or heavily polluted places like gas stations and some specific industries [39].

In the process of decontamination by photocatalysis, the contaminant is absorbed on the surface of the material to be later oxidized, in two stages, to an inert nitrate compound (NO_3). Finally, the inert compound is removed from the surface of the material by rain.

The incorporation of photocatalytic substances in construction materials has emerged as a promising technology to develop products with special characteristics/properties [40]. Among the multiple advantages of photocatalysis is that it is a clean technology that does not need any maintenance, and once applied, its effect is permanent, and it "cleans" the contaminated air. It also saves on costs, since areas where this mortar has been used remain clean for many years, and it destroys the dirt that is deposited on it, which favors the growth of microorganisms [41–45]. Even the possibility to eliminate pollen [46] or deposited soot [47] from the air has been studied.

A building's façade is one of its most important parts since it gives it a distinct personality. On the other hand, pavements in urban areas, steels, parking lots, etc., are among the most important parts of civil construction. In all these cases, it is important to assess the useful life of the chosen material and hence there is a need to investigate new solutions that extend any maintenance operation over time and directly contribute to improving environmental sustainability [38].

With an awareness of the needs of today's society in terms of waste reduction, specifically CDW, through the use of RA and actions on pollution in urban environments, this research aims to contribute to the development of solutions to both problems by developing recycled mortars with photocatalytic capacity that contribute to the preservation of the environment with sustainable initiatives, and are also technically viable. This study investigates the effect of photocatalytic mortars on reducing air pollution produced by traffic emissions of CO_2 and NO_x, with the added value of being made with FRA. The intention is to serve to advance the research work and thus achieve the necessary objectives for sustainable development in accordance with European and Spanish regulations. It contributes to the circular economy by using recycled materials to reduce the waste generated and the need to obtain new raw materials, and improves the quality of life of citizens who live in population centers that have severe pollution problems, which is increasingly present in urban environments.

For this, two families of mortar were manufactured with different types of cement using different replacement rates. The aim was to obtain a recycled mortar that provides the highest decontamination capacity using the highest possible percentage of recycled sand without significantly affecting its mechanical and durability properties.

The rest of the article is structured as follows: the "Materials" section details the materials for the production of the mortars; the "Experimental Program and Methods" section specifies how the research has been carried out; in the "Results and Discussion" section, the data obtained is analyzed; and finally, in the "Conclusions" section, the main advances obtained are highlighted, as well as future lines of research.

2. Materials

The specific materials selected for the research are described below.

2.1. Cements

Two types of cement were used in this research. Cement for the conventional mortar was CEM I 52′5 N (Cement without additions, high strength (52.5 MPa at 28 days), and normal initial strength). The photocatalytic cement used was i.tech ULTRA, hereinafter called Ph. CEM I. This is a Portland cement similar to CEM I, but with an addition of titanium oxide (TiO_2). Both cements come from the same manufacturer.

2.2. Aggregates

This research used two aggregates with a granule size of 0–4 mm:

1. Natural sand (NS): siliceous sand from the Gravera Dehesilla, located in Alcolea (Córdoba, Spain), containing a content of fines (particles < 0.063 mm) equal to 2.26%.
2. Recycled sand (RS): from the Gecorsa CDW treatment plant in Córdoba, Spain, corresponding to the fine fraction of a mixed recycled aggregate; 12.4% of particles were <0.063 mm and it had a solubility in acid equal to 0.87% SO_3. It presented a continuous particle size distribution curve and directly influenced mortar properties such as mechanical strength, workability, compaction, and durability [34].

The composition test carried out on the coarse fraction of said aggregate (>4 mm), according to EN 933-11, showed 18% ceramic particles (Rb), 34% concrete and mortar (Rc), 47% natural aggregate (Ru), and 1% other particles (X), among which plaster stands out. The water absorption and dry specific density of NS and RS, according to EN 1097-6, are shown in Table 1.

Table 1. Specific gravity and water absorption of aggregates. NS, natural sand; RS, recycled sand.

		NS	RS
Dry specific density	(g/cm^3)	2.645	2.573
Water absorption (WA)	(%)	0.32	3.15

The results indicate that RS had a lower specific gravity and higher percentage of water absorption compared with the corresponding properties of natural aggregates, agreeing with what was indicated by other authors [31,35]. This may be due to a higher percentage of mortar and ceramic particles. However, the percentage of water absorption after 24 h of immersion was lower than the 6–9% obtained by other authors [36,48–50]. This may be due to better treatment of the recycled aggregate, or because the original waste contained fewer porous particles.

3. Experimental Program and Methods

3.1. Experimental Program

A total of 8 mortars were manufactured and divided into 2 families, one produced with CEM I (conventional mortar family) that would be used as a reference, and one with Ph. CEM I (photocatalytic mortar family) was used to determine the decontaminating power of mortars made with TiO_2.

In order to increase to the sustainability of the sector, as much RS as possible is expected to be used. For it, each family was produced according to 4 replacement rates of NS by RS (0%, 20%, 40%, and 100%) by weight. Table 2 shows the nomenclature of mortar families produced in this research.

Table 2. Nomenclature of mortar families.

% RS	CEM I	Ph. CEM I
0	M0	PM0
20	M20	PM20
40	M40	PM40
100	M100	PM100

The mortar dosage was calculated based on EN 196-1 and is shown in Table 3. The weights of aggregates shown in this table refer to dry weight.

The amount of mixing water shown in Table 3 was constant, resulting in a water/cement (w/c) ratio equal to 0.58. Due to the low water absorption of RS compared to other recycled aggregates, an increase in the water content as a percentage of increased incorporated RS was not considered, since a high w/c ratio could produce a weaker and more porous mortar [51].

The mixing procedure was in accordance with EN 196-1. A total of 12 prismatic specimens were produced with dimensions of 40 × 40 × 160 mm. These specimens were cured in a climatic chamber at 20 ± 1 °C and 65% relative humidity until the age test.

Table 3. Mortar dosage (per m^3).

	M0/PM0	M20/PM20	M40/PM40	M100/PM100
CEM I/Ph. CEM I (kg)	323	323	323	323
NS (kg)	874	700	525	0
RS (kg)	0	174	349	873
Water (L)	187	187	187	187

3.2. Mortar Characterization

The consistency of the mortar families in the fresh state was measured in accordance with EN 1015-3. The hardened state properties analyzed in the mortar families were compressive and flexural strength (EN 1015-11), water absorption by capillarity (EN 1015-18), water absorption capacity, bulk and skeletal density and open porosity for water (Spanish Standard UNE 8398), carbonation depth (EN 13295), and photocatalytic activity (Spanish Standard UNE 83321 EX). These properties were evaluated after 28 days of curing time.

In addition, X-ray diffraction (XRD) analysis was carried out to identify the main crystalline mineral components. For that purpose, a piece of the central part of each mortar specimen was crushed and sieved through a 0.125 mm sieve. The machine used for this technique was a Bruker D8 Discover A25 with Cu-Kα radiation, and the goniometric exploration used was swept from 5° to 80° (2θ°) at a speed of 0.0142° min^{-1}. The Joint Committee on Powder Diffraction Standards database was used to identify the phases formed in the mortars [52]. A JEOL JMS-7800 scanning electron microscope (SEM) was used to determine the mortar's chemical composition.

For the carbonation depth test, the mortar specimens were introduced into a carbonation chamber under conditions of relative humidity of 55–65%, temperature of 23 ± 3 °C, and CO_2 concentration of 5% ± 0.1%. After 56 days of CO_2 exposure, a phenolphthalein pH indicator spray was used on the mortar fracture surface. The noncarbonated mortar surface showed a purple color due to its high alkaline pH. The carbonation depth was measured on the mortar fracture surface from the edge of the specimen to the purple area.

Despite the successful application of TiO_2 photocatalysis to cement-based materials, an ideal method to determine the photocatalytic activity is still not available. The experimental conditions and data treatment differ in many aspects (light source, UV intensity, temperature, humidity, flow rate, characteristics of test samples, contaminant analyzed), even leading to noncomparable results [42]. The experimental method proposed by Spanish standard UNE 83321 EX was used in this research. This was aimed at evaluating the degradation of nitrogen oxide, in the gas phase, of inorganic photocatalytic materials contained in cement concretes by a continuous flow test method. For the measurements and calculations required in this test, the concentration of nitrogen oxides (NO_x) was defined as the stoichiometric sum of nitrogen oxide (NO) and nitrogen dioxide (NO_2).

Likewise, a sample was extracted from the center of each prismatic specimen mortar to analyze its photocatalytic power according to the standardized methodology through the reduction capacity of NOx. The test was carried out on the photocatalytic mortar family in addition to the reference mortar (M0).

4. Results and Discussion

4.1. Consistency

The average consistency results obtained after two perpendicular measurements are shown in Table 4. It can be seen that as the percentage of RS increases, the consistency of the mortar decreases, with a minimum value corresponding to 100% RS, mainly due to its higher water absorption. Silva et al. [38] stated that this loss in consistency can also be attributed to the greater angularity of the recycled particles, avoiding effective slippage between them. To compensate for this, an amount of water corresponding to the absorption could be added [31,36,49,53,54], although the mechanical properties could be affected [35]. For this reason, the most suitable solution would be to add plasticizers to the mortar mix, thus increasing its consistency/workability, as advised by Ledesma et al. [14].

Table 4. Consistency values of mortar families (mm).

	Consistency		Consistency
M0	134	PM0	129
M20	131	PM20	125
M40	130	PM40	113
M100	111	PM100	102

It was also observed that the photocatalytic mortar family showed less consistency in all cases compared to the conventional family. The lowest consistency, registered by PM100, may be responsible for its porous appearance (Figure 1) and difficult compaction.

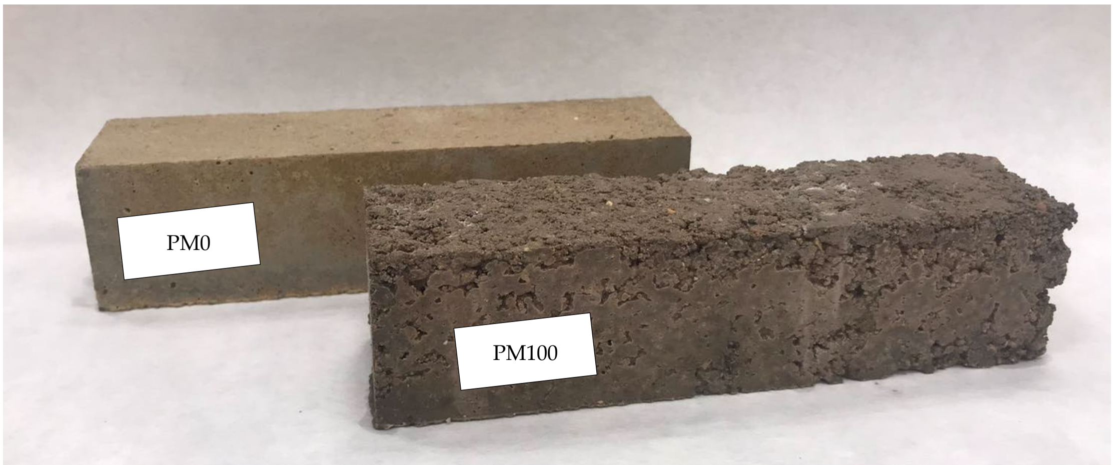

Figure 1. Porous appearance of PM100 versus smooth appearance of other mixes.

4.2. Mechanical Strengths

The values registered for mechanical strength at 28 days are shown in Figure 2, which illustrates the comparison of results between the two families.

Figure 2a shows that for the family made with conventional cement and 40% of NS replaced by RS (M40), the compressive strength is equal to that corresponding to 20% replacement, approximately 46 MPa, which is only 10% less than the reference mortar (M0) at 52 MPa. For full replacement (M100), the drop in compressive strength increases to 16%, with an average value of 44 MPa. This agrees with Silva et al. [51], suggesting that as the RS content increases, the compressive strength remains similar to or larger than that of the control mortar. This may be due to the reduction of effective water as the percentage of RS is increased, as explained by López Gayarre et al. [35], or because of the greater number of fine particles that can fill the gaps [49]. In this investigation, the content of fine RS was higher than NS (12.4% vs. 2.26%), favoring the filling of voids in the mortar matrix and diluting the loss of mechanical strength. However, in most mortar mixes, there is a loss of compressive strength as the replacement percentage increases [34]. In all mortar mixes, the compressive strength is greater than the value recommended by the GB 28635-2012 standard [55] (average strength ≥ 30 MPa; any individual strength ≥ 25 MPa) and values obtained in other studies [19,56].

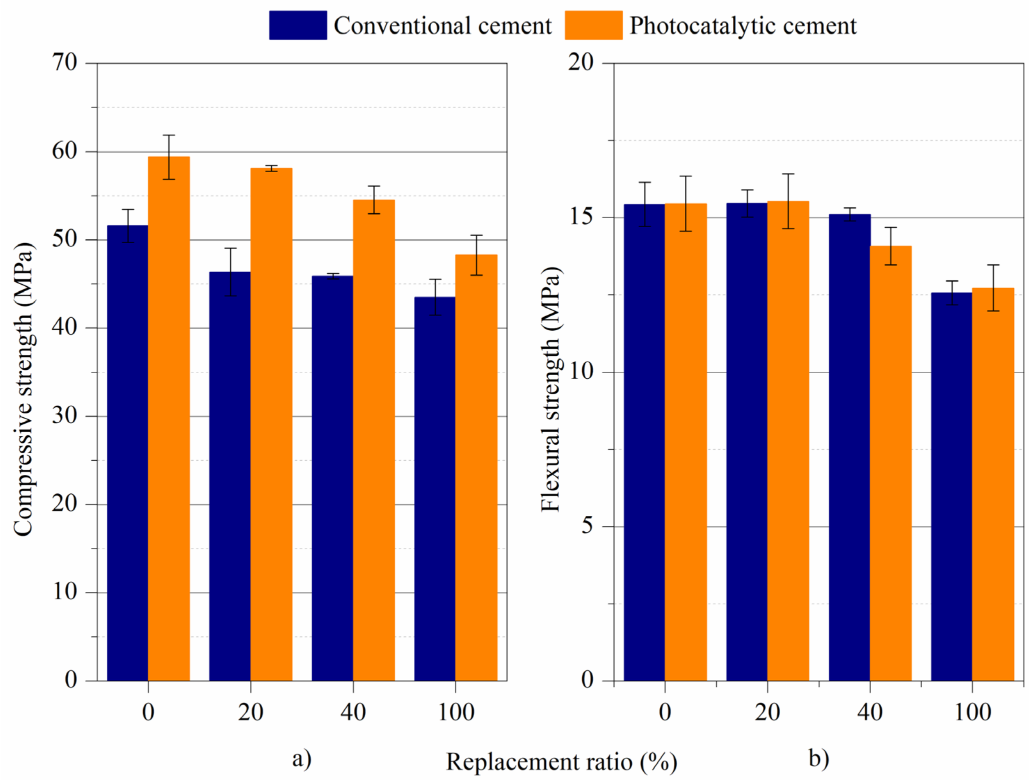

Figure 2. Mechanical strength comparison: (**a**) compressive strength; (**b**) flexural strength.

In all cases, the compressive strength obtained in the photocatalytic family is greater than that of their counterparts made with conventional cement. In this way, they start from an average resistance of 59 MPa for PM0, reaching 48 MPa for PM100.

Flexural strength can be correlated with other characteristics such as susceptibility to cracking and adhesive strength of mortar [51]. In this test (Figure 2b), a similar value of flexural strength was obtained for the 0%, 20%, and 40% replacement, 15 MPa, except for the M100 and PM100 families, for which a value of 13 MPa was found. This agrees with values obtained by Silva et al. [51], whose research used the same type and dosage of cement. Therefore, no improved behavior of the photocatalytic family was observed in this test. All values registered in the flexural strength test show very good performance of these mortars for their possible use in pavement, with values greater than 12 MPa in all mortars, exceeding the values obtained by other authors [56,57].

The differences between the strength of mixtures with 0% and 40% RS are practically insignificant, which confirms that increasing the percentage to 40% does not just mean decreased mechanical properties. This agrees with Ledesma et al. [14], who established a maximum replacement ratio of up to 50% of natural sand by mixed recycled sand without significantly affecting the hardened mortar properties.

Generally, a decrease in strength is observed as the percentage of recycled aggregate is increased, as reported in previous works. This decrease is more gradual and more noticeable in the photocatalytic family. However, the increase in RS up to 40% showed only a 7% decrease in strength with respect to 20% replacement, and up to 9% with respect to the reference mortar.

4.3. Mineralogical Analysis

The mineral phases formed in conventional and photocatalytic mortar families are shown in Figures 3 and 4, respectively. In all specimens, the main detected phase corresponds to quartz (SiO_2; 33-1161) [52]. The intensity decreased in all specimen patterns because the NS was replaced with RS, which contains less silica [58]. The main phase in M100 was sanidine ((Na, K)(Si_3Al)O_8; 10-0357) [52].

The Portlandite phase ($Ca(OH)_2$; 04-0733) [52] and ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$; 41-1451) [52] were also observed in both families. The presence of these phases is an indicator of the Portland cement reaction [57], as shown by the mechanical performance of both mortar families (Figure 2). For this reason, the incorporation of RS into the mortar is compatible with the common Portland cement reaction.

The other detected phases in the mortars corresponded to silicates such as illite ($KAl_2Si_3AlO_{10}(OH)_2$; 02-0056) [52] and albite ($Na(Si_3Al)O_8$; 10-0393) [52], which is in agreement with authors such as Jiménez et al. [36] and Ledesma et al. [14]. Regarding carbonates, all mortars showed calcite ($CaCO_3$; 05-0586) [52]; additionally, dolomite ($CaMg(CO_3)_2$; 36-0426) [52] was detected in M100 and all specimens of the photocatalytic mortar family. In both mortar families, it was observed that calcite ($CaCO_3$; 05-0586) [52] had a greater presence as the replacement of NS by RS increased. This behavior has also been recorded by authors such as Gonçalves et al. [58], who studied the replacement of natural siliceous sand with recycled aggregate. Gypsum ($CaSO_4\cdot 2H_2O$; 33-0311) [52] was also detected in all specimens in both families, which could correspond to the RA or cement composition [36].

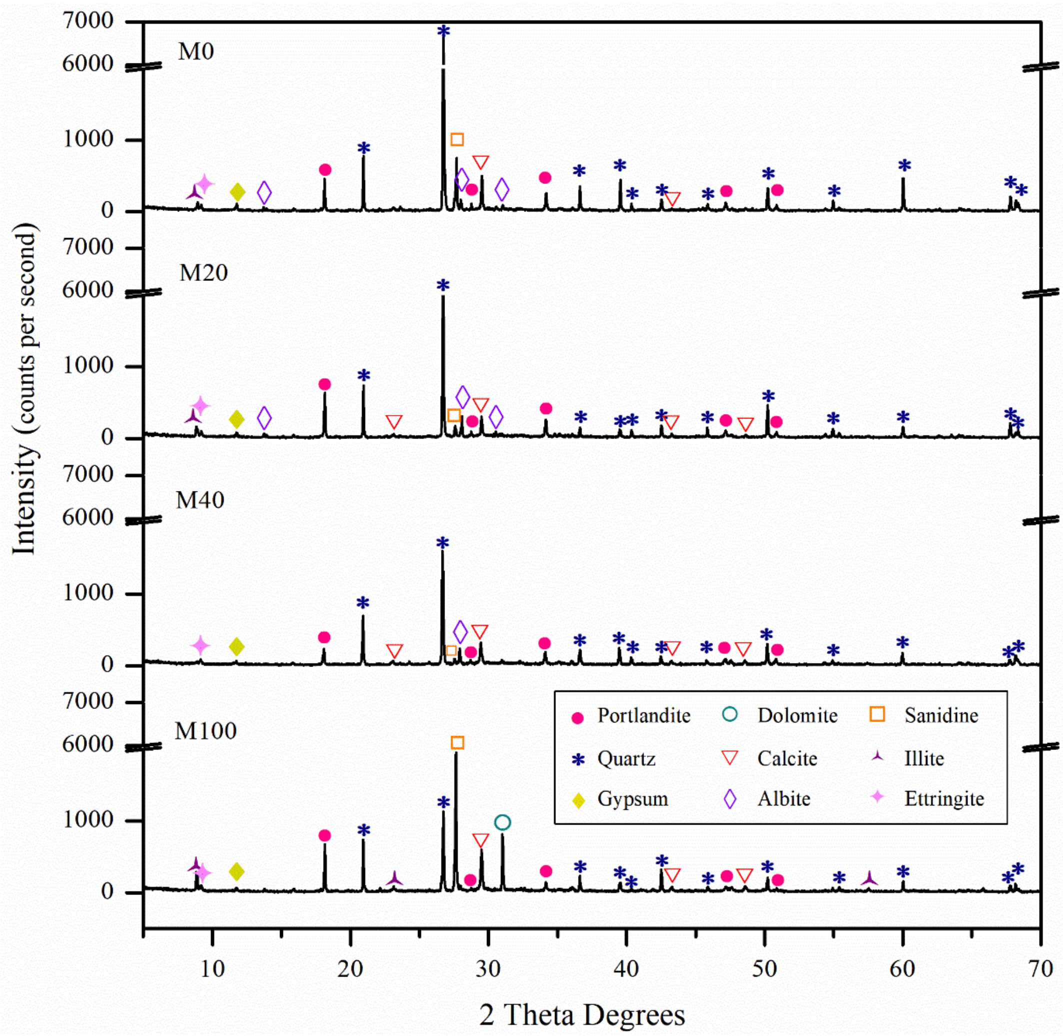

Figure 3. X-ray diffraction patterns of conventional mortar family.

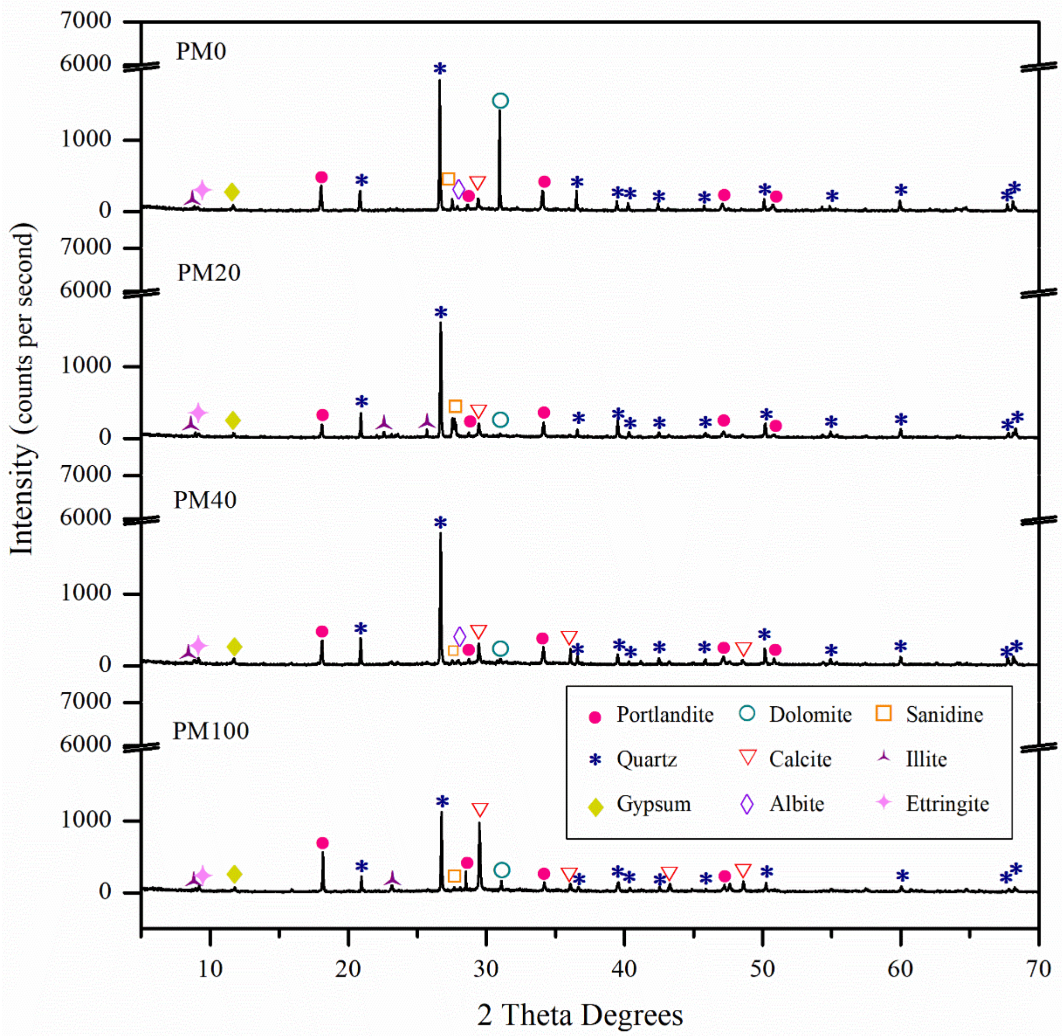

Figure 4. X-ray diffraction patterns of photocatalytic mortar family.

Additionally, an SEM study was carried out, which supported the results obtained in the mineralogical study, as shown in Table 5. In the photocatalytic mortar family, no mineralogical phase attributed to Ti was detected; however, SEM confirmed the presence of TiO_2 in all mortars with Ph. CEM I.

Table 5. Average percentage, by weight, of elements by scanning electron microscopy (SEM).

Element	M0	M20	M40	M100	PM0	PM20	PM40	PM100
C	10.30	8.53	9.31	9.40	7.79	12.15	10.42	12.16
O	49.65	48.59	50.10	45.34	48.79	46.67	45.84	49.39
Mg	0.50	0.87	0.52	0.56	0.31	0.37	0.30	0.51
Al	2.30	2.95	2.36	3.89	1.78	3.00	3.74	1.96
Si	8.01	9.72	8.27	10.61	9.46	13.78	10.61	12.18
S	0.75	0.95	1.14	0.92	0.62	0.66	0.75	0.90
K	0.34	0.39	0.41	1.06	0.71	0.32	1.64	0.39
Ca	26.34	25.58	26.51	25.14	28.96	20.40	23.69	20.48
Ti	0.00	0.00	0.00	0.00	0.52	0.55	0.63	0.63
Total:	100	100	100	100	100	100	100	100

4.4. Water Absorption by Capillarity

The capacity to absorb water indicates the ability of an unsaturated porous material to absorb and drain water by capillary action, thus making it a suitable property to indirectly assess the durability of cementitious materials. Normally, higher water absorption by capillarity contributes to worse performance since it impairs the protection against external agents [51].

The results shown in Figure 5 indicate an upward trend in water absorption by capillarity as the percentage of RS increased in both families. However, for 20% of RS (M20), the values achieved in the conventional mortar family were even lower than those of the reference mortar (M0). The higher water absorption by capillarity of recycled mortars can be due to the high absorption of RS. Similar tendencies have been observed in studies by other authors [14,31,34,36].

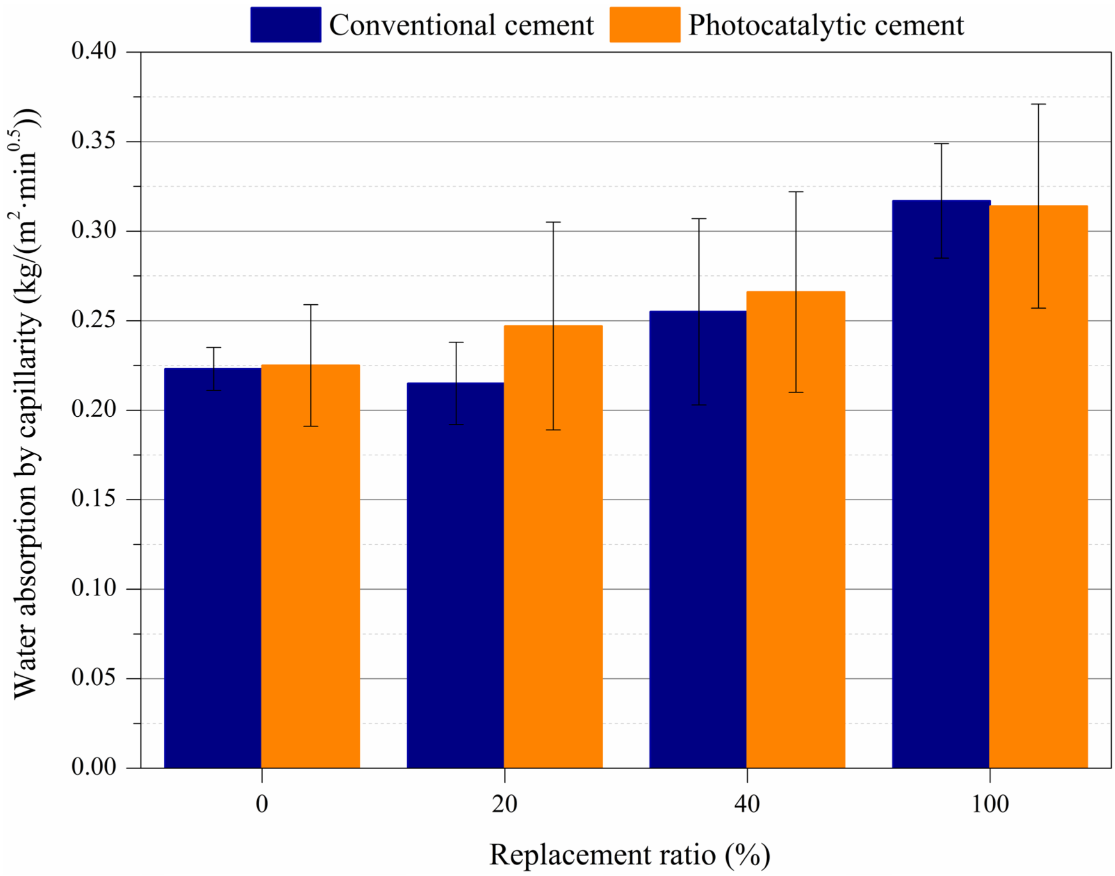

Figure 5. Water absorption by capillarity comparison.

According to López Gayarre et al. [35], the reduction in the amount of effective water as the percentage of substitution is increased (greater water absorption) reduces the porosity of fresh mortar, and for this reason, recycled mortar could present this slight increase of water absorption by capillarity.

4.5. Water Absorption Capacity, Bulk and Skeletal Density, and Open Porosity

The water absorption capacity (Figure 6a) was almost the same for all mortars produced with photocatalytic CEM I. However, for the conventional mortar family, there was a slight increase with 40% of RS, and a more appreciable increase for 100% of RS of around 25%.

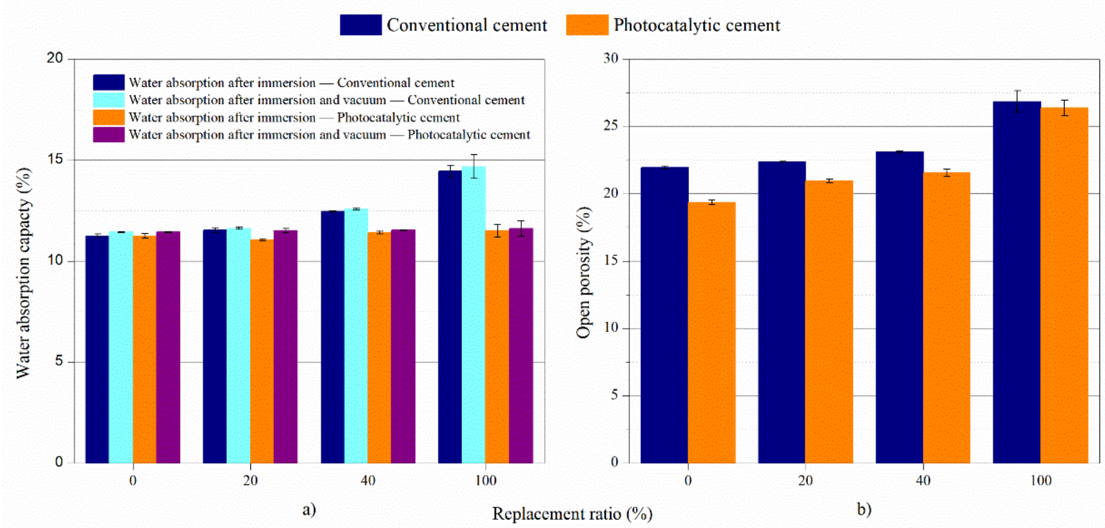

Figure 6. Comparison of results for (**a**) water absorption capacity and (**b**) open porosity.

Open porosity (Figure 6b) was higher as the RS replacement ratio increased. Poon and Cheung [59] explained this, affirming that materials with lower density lead to higher porosity of mortar blocks.

However, the density values (Figure 7a,b) were approximately the same in all cases, regardless of the type of cement or the amount of recycled aggregate. This differs from the values obtained in some studies [31,34], in which the density decreased as the replacement ratio increased. Other studies [31,49,60,61] found no significant differences in replacement ratios below 20–25%, while for higher replacement ratios, the lower dry density of FRA decreased the dry density. This result was attributed to the fact that a higher fine content (<0.063 mm) in RS allows for filling of voids at replacement ratios up to 10% of the hardened mortar.

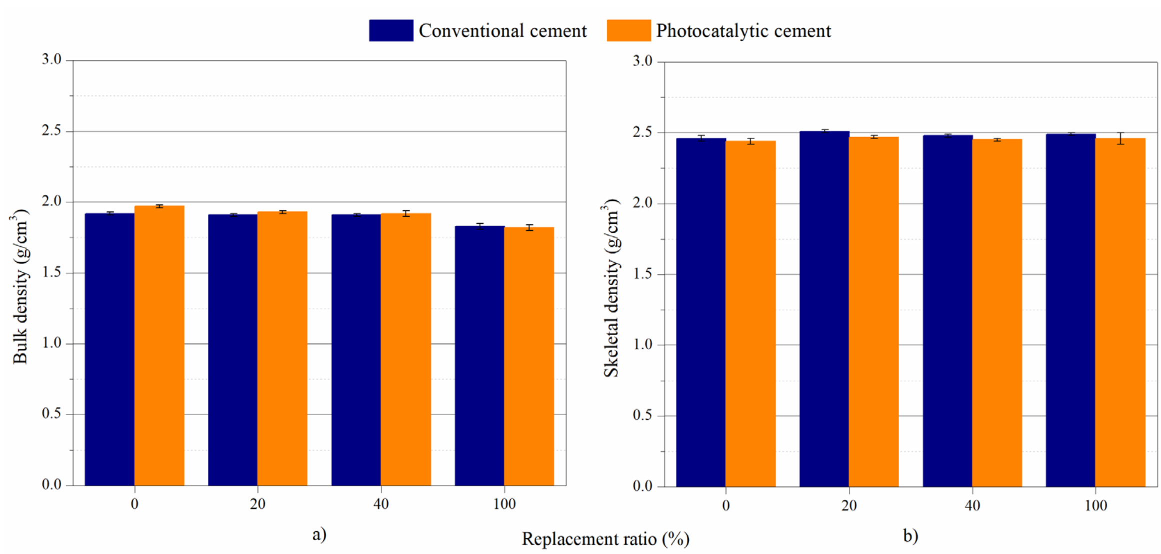

Figure 7. Comparison of results of (**a**) bulk density and (**b**) skeletal density.

4.6. Carbonation Depth

The results of the carbonation test are shown in Figure 8. The carbonation depth was greater when the RS increased, although this increase was slight, up to 40%. For 100% replacement, the increase in carbonation depth was much greater than in the reference mortar, possibly due to the greater porosity of this mixture, as observed in the previous section.

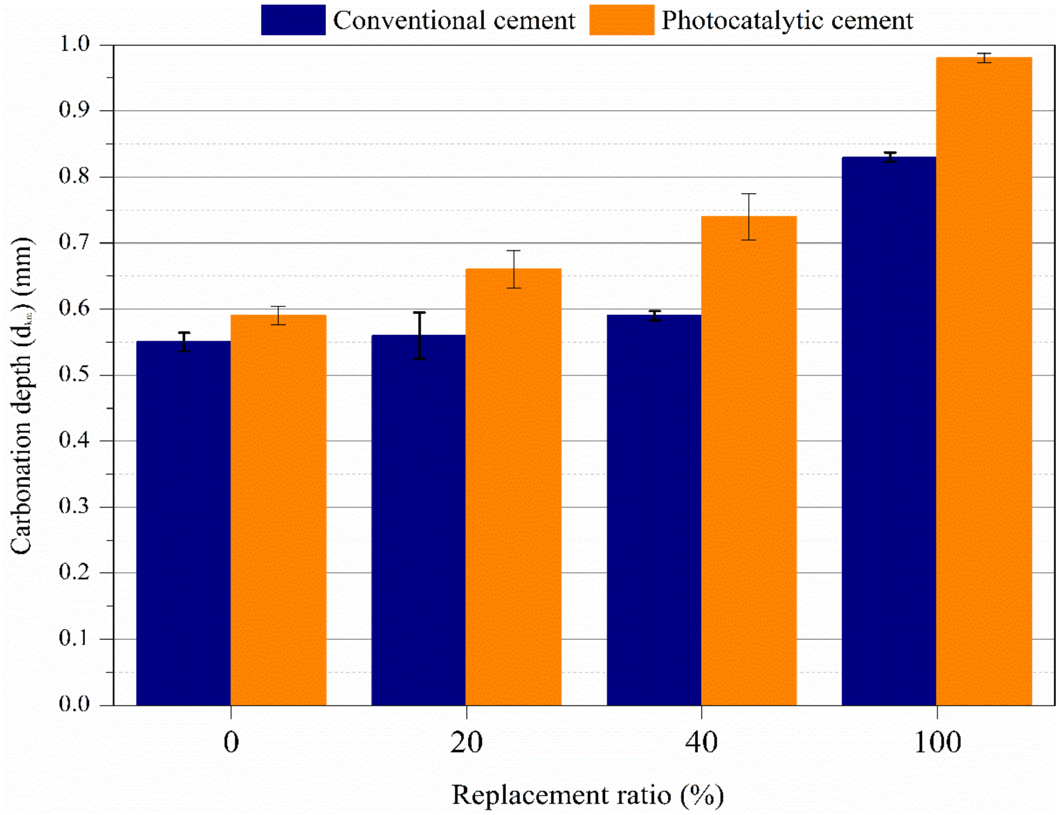

Figure 8. Carbonation depth comparison at 56 days.

This behavior was similar in both families, although it was more appreciable in the photocatalytic mortar family. Specimens of the photocatalytic mortar family showed an increase between 7% and 25% (17% average) higher than their counterparts in the conventional mortar family. This property showed the same trend as reported by Moro et al. [50].

Of all the mortars, PM100 is the one with the highest CO_2 absorption, with a 78% increase in carbonation depth as compared to the reference conventional mortar (M0), and a 66% increase as compared to the reference photocatalytic mortar (PM0). Thus, there is an added beneficial effect for the environment, since carbonation involves the absorption of CO_2 from the air. This reacts with the $Ca(OH)_2$ from cement hydrolysis, producing CO_3Ca and immobilizing CO_2. This process is detrimental to reinforced concrete, since it greatly affects durability due to the risk of corrosion of reinforcements by reducing the pH of the concrete. However, since mortars are not armored, that would not be a problem.

4.7. Photocatalytic Activity Test

According to the results of Figure 9, incorporating RS instead of NS slightly improved the decontaminating capacity. This can be explained by the greater porosity of the mortar as the percentage of recycled sand was increased, as observed in the previous sections, and agrees with the results reported by Poon and Cheung [59].

According to Spanish Standard UNE127197-1, most of the mixes were classified as category 1; that is, with decontaminating power, measured as the reduction of NO_x varying between 4$ and 6% (4.2%, 4.3%, and 4.8% for PM0, PM20, and PM40, respectively). However, the mix made with photocatalytic cement (PM100) and 100% RS showed a porous appearance and greater decontaminating capacity within category 2 (7.2%), which represents an increase of 71% of decontaminating power compared to mortar made with NS (PM0). This is consistent with Poon and Cheung [59], who indicated that the porosity of the surface layer is important, as it effectively increases the area available for reacting with pollutants.

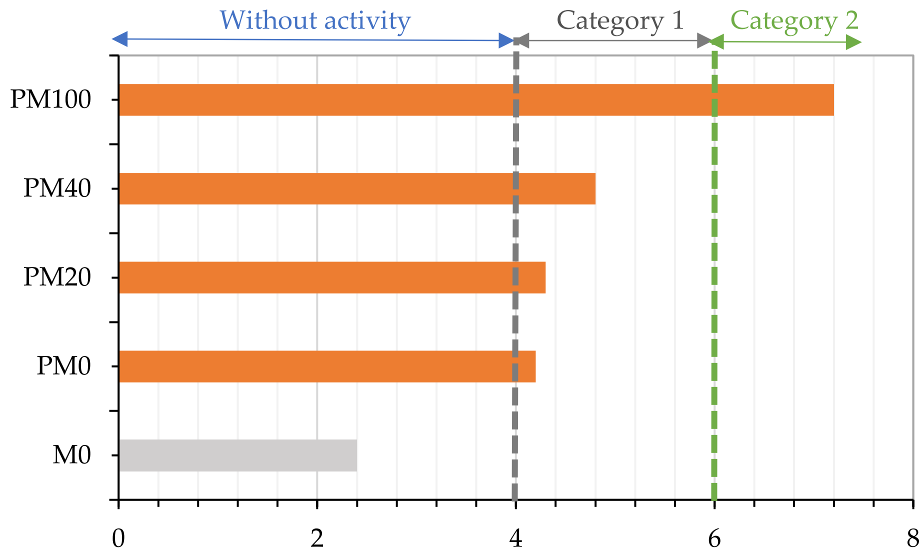

Figure 9. Elimination of NO_x (%).

5. Conclusions

This research produced mortars with decontaminating capacity by introducing waste into the productive cycle and contributed to the implementation of the circular economy model. The results show suitable mechanical behavior despite the incorporation of recycled aggregates, since total replacement of NS by RS meant a decrease in compressive and flexural strength of only approximately 18%, with average values of 46 MPa and 13 MPa, respectively. The strength obtained with photocatalytic cement was slightly higher compared to its counterpart made with traditional cement. The properties of water absorption by capillarity, water absorption capacity, and open porosity showed a slight increase for 40% and 100% RS, while in the results obtained for 20% RS, the values obtained were very similar to those of the reference mortar. The bulk and skeletal density showed very similar values in the two families and for all replacement ratios.

The penetration of CO_2 obtained in the carbonation test carried out showed a clear benefit with the incorporation of recycled sand. This could be a positive aspect, as it reduces the carbon footprint in the environment.

From a photocatalytic point of view, the incorporation of up to 40% RS slightly favored the elimination of NO_x, but the mortar with 100% RS had a significant increase relative to the conventional mortar, moving to a better classification (category 2). These mortars were more porous than conventional mortars, facilitating the entry of light into the interior and, consequently, the elimination of polluting gases. The results obtained add value to the use of recycled aggregates, clearing up the uncertainties that still exist in the use of this material.

Therefore, a photocatalytic mortar with 100% mixed recycled sand is proposed in order to produce the greatest environmental benefits, due to the greater absorption of CO_2 and NO_x and the greater use of recycled aggregates. Also, the strength is slightly lower but compatible with its use in low-requirement applications in contact with the atmosphere, such as pavement blocks or cladding mortar.

The findings of the present study prove that reducing natural sand mining, minimizing energy consumption and CO_2 emissions, reducing global warming, preventing illegal deposition and landfilling of the fine fraction of CDW, and complying with the limits of the European Waste Framework Directive are possible in order to achieve and promote cleaner production in the construction sector (eco-efficiency).

Future lines of research are intended to improve the photocatalytic capacity of mortars based on the good mechanical behavior of the mortars studied in this research by studying the influence of the content and origin of fine aggregates, different aggregates and their nature, reducing the amount of cement due to the good mechanical behavior obtained, or increasing the amount of water according to the percentage of increased RS, or pre-saturating it.

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