



Research article

Environmental assessment of alkali-activated materials based on agro-industrial waste as alkaline activators through leaching tests

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ABSTRACT

Global circular economy drives the development of sustainable alkali activated materials (AAM) for use as construction material from industrial by-products and wastes. The assessment of the potentially hazardous substances release of these new material combinations into the soil and groundwater over time is essential. In this study, the aim is the environmental assessment of three AAMs based on blast furnace slag (BFS), activated with almond shell biomass ash (ABA) as potassium source and three solid sources of silica from the agricultural industry, rice husk ash (RHA), spent diatomaceous earth (SDE) and bamboo leaf ash (BLA), using European horizontal leaching tests proposed for construction materials, for monolithic form, Dynamic Surface Leaching Test (DSLTL) and for granular form, Up-flow Percolation Test and the Compliance leaching test, by simulating different scenarios of their entire life cycle. The leaching results of the AAM showed the effectiveness of the inertization of all the recycled materials studied, which exceeded some inert materials limits, by means of the activation process. Despite the absence of significant differences in the leaching mechanisms of the oxyanions As, Cr, Mo, Sb, Se and V between the three AAMs developed, they presented different long-term leaching behavior depending on their form, monolithic, or granular, and therefore in their different life cycle stages. Therefore, it is concluded that although the incorporation of agro-industrial waste as alternative activators in BFS based AAM according to the Dutch Soil Quality Decree (for unrestricted use of monolithic and granular materials) is an environmentally acceptable option, the design of waste derived AAMs should be assessed by means of a combination of leaching tests that cover their expected life cycle.

1. Introduction

Alkali-activated materials (AAM) are a promising alternative to construction materials manufactured with Portland cement, achieving similar performances in diverse applications with lesser embodied CO₂ due to the absence of calcination of limestone and combustion for clinker production (Provis and Bernal, 2014). Alkali-activated materials or geopolymers are comprised of aluminosilicate precursors, that are mainly based on secondary resources, and activators, thus significantly reducing their environmental impact and avoiding the disposal of industrial waste streams in landfills, which is in line with the adoption of circular economy practices (Nodehi and Taghvaei, 2022). This is why research interest about AAM is being severely increased, and recent advances are focused towards finding secondary raw materials supply to

have an abundance of options in order to manufacture AAM worldwide (Palomo et al., 2021), thus increasing the available resources and further decreasing their cost and environmental footprint. In particular, commercial activators commonly used in the manufacture of AAM such as sodium and potassium silicates have the highest economic cost and environmental impact derived from their production (Alnahhal et al., 2021; Yu et al., 2023), since the calcination (1400–1500 °C) of sodium/potassium carbonate and siliceous sand entails a significant release of CO₂ (Alnahhal et al., 2023). Hence, significant environmental improvement in the manufacture of AAM can be achieved if commercial activators are substituted by secondary resources (Kumar et al., 2023).

In this line, industrial wastes and biomass ashes are of great interest to substitute commercial activators because they constitute a widely available source of materials in quantity and variety (Kumar et al., 2023;

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Payá et al., 2022). They can be divided into silica rich and alkali rich activators. The silica rich materials may substitute sodium or potassium silicate while wastes that are high in sodium or potassium oxides can serve as alternatives to alkaline sodium/potassium hydroxide (Soriano et al., 2020). Some of the authors of this study have demonstrated the technical feasibility of AAMs containing agro-industrial waste-based activators rich in both silica and alkali when synergistically used with blast furnace slag (BFS) as precursor in previous works (Font et al., 2020; Soriano et al., 2022). However, their environmental impact assessment has not been addressed yet.

Construction materials can pose a significant impact on the local environment, as they interact with groundwater, surface waters, marine waters, or soil and may release potentially harmful substances through leaching processes (van der Sloot et al., 2017). The leaching assessment of materials combinations based on waste or by-products should be carried out to check their potential environmental risks from a circular economy perspective (Engelsen et al., 2021; Ferrazzo et al., 2023), and therefore it is necessary to assess the risk of these materials in their life cycle.

In this context, the European Construction Products Regulation indicates “hygiene, health and environment” as a category of basic requirements for construction works to comply during their entire life cycle (Bandow et al., 2018). Two different leaching test procedures have been proposed to assess the release of substances into water. The Dynamic Surface Leaching Test (DSLTL) was created to study the leaching from the surfaces of monolithic construction products, as a function of exposed area and time; and the Up-flow percolation test that is applied to assess the release of granular construction products, as a function of the liquid-to-solid ratio. The results of both tests can help to deduce the mechanisms governing the leaching of each substance (García-Ruiz et al., 2020).

Until now, only a few studies on leaching behavior of waste-derived AAM or geopolymers using one of these European standard tests have been published, specifically the DSLTL to monolithic samples (Maldonado-alameda et al., 2021; Sun et al., 2022; Sun and Vollpracht, 2020) and the Up-flow percolation test to granular samples (Gijbels et al., 2019). In these studies the substitution of commercial precursors by waste was explored, while the leaching behavior of AAM that include alternative activation sources such as rice husk ash (RHA) (Fernando et al., 2022; Guo et al., 2024) and olive biomass ash (OBA) (Alonso et al., 2019) have been addressed with single-stage or batch tests to evaluate the compliance of the material with regulatory leaching levels. Batch equilibrium tests are designed to represent the release of substances under different conditions by changing a single parameter, such as the liquid-to-solid ratio (L/S) or the final pH of the leachate (Engelsen et al., 2021), which has been demonstrated to be insufficient to assess the environmental impact in the different stages of their life cycle of these type of materials (Kumar et al., 2023).

As far as the authors know, to date only a few studies have focused on this aspect of waste-derived activators (Wu et al., 2024), and the environmental assessment through a combination of leaching tests has not been addressed, although it is considered essential, considering the severity of the risks of leaching of potential contaminants into the environment in the new context of materials circularity. Therefore, the aim of this study is to focus on the environmental assessment of AAM that incorporate agro-industrial by-products as substitutes of commercial activators using leaching criteria. The AAM explored herein are completely comprised of industrial by-products; BFS activated with almond shell biomass ash (ABA) as alkali source based on potassium and three different silica sources that were used to replace commercial sodium silicate, rice husk ash (RHA) from the rice industry, spent diatomaceous earth (SDE) from filtration processes of the brewery industry and bamboo leaf ash (BLA) from a bamboo cane company. The materials developed were tested through a battery of European harmonized leaching tests, Compliance leaching test (EN 12457-4), Dynamic surface leaching test (CEN/TS 16637-2), and Up-flow percolation test (CEN/TS

16637-3). These tests cover the expected release of elements of potential concern during their overall life cycle, from production, through the period of use, to demolition and reuse (as secondary material) or disposal (as a waste).

The novelty of this study is the environmental research on innovative valorization of waste and industrial byproducts as alternative activators into AAM formulations using leaching criteria throughout their life cycle. The leaching results and mechanisms obtained allow to predict the long term impact of these materials in the different scenarios and thus validating their potential uses by different stakeholders.

2. Materials and methods

2.1. Raw materials

All the secondary raw materials employed in the formulations of AAM consisted in recycled materials from different industrial processes. The precursor used in the present investigation was the BFS, supplied by Cementval S.A (Puerto de Sagunto, Spain). The ABA supplied by Borges Agricultural & Industrial Nuts (Altura, Spain) was used as alkali source based on potassium. Three different sources of silica were used to substitute the commercial sodium silicate, these materials were: i) RHA supplied by Dacsa S.A (Tabernes Blanques, Spain); ii) SDE from wine industry supplied by Bodegas Vicente Gandía (Chiva, Spain); iii) BLA produced by an auto combustion process (Ilha Solteira, Brazil).

2.2. Alkali-activated mortars preparation and characteristics

The AAM were designed to include the largest proportion of BFS, as more precursor is needed than activator in the alkaline activation systems. To do this, the doses as percentage of binder weight were 75% BFS, 12.5% ABA and 12.5% of each silica source (RHA, SDE and BLA). Three AAM mixtures were developed and named accordingly to the type of silica source included; alkali-activated materials using RHA, SDE and BLA as silica source are labelled as AAM-RHA, AAM-SDE and AAM-BLA, respectively. All materials were ground together in a ball mill for 30 min to obtain the one-part mixtures with an average particle diameter of 21 μm to ease the alkali activation process.

The milled material was used to prepare mortars with an AAM/sand proportion of 1/3 and a water/binder ratio of 0.45. The mortars were obtained as one-part mixtures, based on the procedure used to mix sand with cement UNE-EN 196-1 (UNE, 2018). The mortars were cured at room temperature (20 °C and 90 % humidity) until age of mechanical test. Four values for the compressive strength of specimens (4 × 4x4 cm³) were obtained according to UNE-EN 196-1 (UNE, 2018) for each mixture using a Ibertest MUP-60 apparatus. The dosage of mortars is shown in Table 1.

The compressive strength of all AAM measured at 7, 28 and 56 curing days at ambient temperature is depicted in Fig. 1.

The combination with SDE had the best mechanical performance at all curing ages. In general, the values obtained with the AAM with SDE and BLA are close to the value obtained by a cement with a category of 42.5 MPa at 28 curing days (Soriano et al., 2022). While in the literature, for one-part geopolymers using metasilicate as commercial activators prepared at ambient temperature, values between 49.2 MPa (Zhang

Table 1
Mix design of the AAM.

Materials	BFS	ABA	RHA	SDE	BLA	Sand	Water
AAM-RHA (kg m ⁻³)	337.5	56.25	56.25	–	–	1350	202.5
AAM-SDE (kg m ⁻³)	337.5	56.25	–	56.25	–	1350	202.5
AAM-BLA (kg m ⁻³)	337.5	56.25	–	–	56.25	1350	202.5

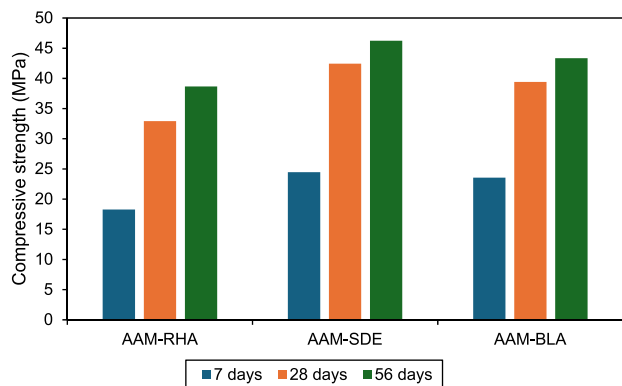


Fig. 1. Compressive strength of AAM mortars cured at ambient temperature at 7, 28 and 56 days.

et al., 2021) (mixture of fly ash and metakaolin as precursor for 7 days curing time) and 20–30 MPa (Shoaei et al., 2024) (mixture of BFS and a natural zeolite in different proportion for 28 days curing time), are reported. Therefore, the compressive strengths obtained in this study of AAM using waste-derived activators can be considered acceptable.

2.3. Leaching tests

2.3.1. Compliance leaching test

The test is performed according to EN 12457-4 (European Committee for Standardization, 2003). As other batch tests (e.g. TCLP, ASTM D3987-12), they intend to reach chemical equilibrium between a granular solid and a leachant (deionized water) (Moghal et al., 2023). Raw materials and crushed AAM with a particle diameter lesser than 10 mm were tested by batches, weighting 90 ± 5 g of sample in 1 L plastic bottles. 900 mL of deionized water were introduced, obtaining a liquid-to-solid ratio (L/S) of 10 L/kg. Immediately after, the bottles were introduced in a rotary shaker (Heidolph Reax 20) with a rotation speed of 10 rpm during 24 h. After this agitation period, samples were filtered in a glass equipment through a 0.45 μm Millipore membrane filter, and pH and conductivity were measured. Leachates were divided into two aliquots, one acidified with analysis grade HNO_3 to a $\text{pH} < 2$ and other without acidification and both aliquots were stored at 4 ± 2 °C prior to chemical analysis. Test was performed in triplicate for both raw materials and AAM.

2.3.2. Dynamic surface leaching test (DSLTL)

The test was performed according to the technical specification CEN/TS 16637-2. Cubic monolithic AAM samples of $4 \times 4 \times 4$ cm³ of dimensions were statically suspended in a 1 L plastic vessel with a previously washed nylon net, in order to assure exposure of all the faces, with at least 2 cm of distance between the walls of the vessel. The test consisted of 8 successive stages in which deionized water was replaced in a liquid to exposed area ratio of 80 L m⁻². In each stage, 780 mL of deionized water were introduced and immediately after the vessel was closed in the upper part with a plastic lid to avoid carbonation and external interferences. The collection of leachates took place in predefined intervals of time, at 0.25, 1, 2.25, 4, 9, 16 and 36 days since the start of the test, with a last collection at 64 days. The leachate collected in each stage was measured its pH and conductivity, filtered, acidified with HNO_3 to a $\text{pH} < 2$, and stored prior to chemical analysis. The test was performed in triplicate.

2.3.3. Up-flow percolation leaching test

The up-flow percolation leaching test CEN/TS 16637-3 for granular construction materials was applied to the AAM. Samples were crushed and sieved to a maximum particle diameter of 16 mm. Grounded

samples were introduced and compacted in polycarbonate columns of 300 mm of height and 50 mm of diameter. A multichannel peristaltic pump was employed to force the flow the leachant (deionized water) upwards through the columns. Prior to test start, columns were saturated with leachant and left 72 h to reach chemical equilibrium state between solid and leachant. After that time, leachant was pumped at a constant flowrate of 300 mm/day. Extractions of leachates were performed in 7 stages at predefined cumulative L/S ratios: 0.1, 0.2, 0.5, 1, 2, 5 and 10 L/kg. Immediately after collection, pH and conductivity of the leachates were measured, filtered, acidified with HNO_3 to a $\text{pH} < 2$ and stored for chemical analysis. Tests were performed in duplicate.

2.4. Analytical methods

The chemical composition of raw materials as major oxides and trace elements content was determined by lithium metaborate-tetraborate fusion and total digestion with inductively coupled plasma optical emission spectroscopy (ICP-OES) and Instrumental Neutron Activation Analysis (INAA) in the Actlabs facilities in Ancaster (Canada).

The mineralogical analysis of the materials used to prepare the AAM was performed in an X-ray diffractogram (XRD) using a Bruker AXS D8 Advance. The voltage used was 40 kV and the current intensity was 20 mA. The diffractograms were recorded for the 2 θ range between 5° and 70°, with a steep angle of 0.02° and an accumulation time of 2 s.

Trace elements concentration in leachates were analyzed with an Agilent 7900 \times Inductively Coupled Plasma Mass Spectrometer (ICP-MS), and anions Cl^- , F^- and SO_4^{2-} were determined through ion chromatography in a Compact Ion Chromatograph Flex 930 (Metrohm), in the Campus Terra facilities of the University of Santiago de Compostela (Spain) according to ISO quality control standards. Quantification limits for the trace elements considered were ($\mu\text{g L}^{-1}$): As (0.033), Ba (0.61), Cd (0.007), Cr (0.063), Cu (0.02), Hg (0.042), Mo (0.03), Ni (0.027), Pb (0.018), Sb (0.003), Se (0.039), V (0.018) and Zn (0.42), and for the anions ($\text{mg}\cdot\text{L}^{-1}$): Cl^- (1.5), F^- (0.02), SO_4^{2-} (2.5).

3. Results

3.1. Characterization of raw materials

In AAM systems, precursors and activators that may contain potentially hazardous trace elements are subjected to changes in their chemical structure as a consequence of alkali activation reactions, conforming new compounds that may reduce or enhance the mobility of these trace elements. Therefore, it is crucial to have knowledge of the composition and mineralogy of the starting materials prior to inclusion in the AAM. The chemical composition of the different raw materials is shown in Table 2.

The BFS is rich in SiO_2 and CaO , with minor presence of Al_2O_3 and MgO , and ABA is mainly composed of K_2O and CaO . The three alternative materials considered to replace commercial silicates, RHA, SDE and BLA, have a silica oxide content superior to 70%, with minor contents of K_2O in RHA, Al_2O_3 in SDE, and Fe_2O_3 and Al_2O_3 in BLA. Attending to the trace elements content, overall high levels are found in the ABA and SDE (Cd, Cr, Cu, Mo, Ni, Pb, Sb, V), while others are higher in RHA (As, Hg), BFS (Ba) and BLA (Cr, V, Zn).

In Fig. 2 the diffractograms of the secondary raw materials are represented. All phases determined are in agreement with the major chemical composition of the materials. The presence of amorphous phases is observed by the baseline deviation in the interval of 2 theta (degrees) between 5° and 35°. The diffractograms of all raw materials present this deviation, but in BLA (Fig. 2 (e)) is less visible. In BFS (Fig. 2 (a)), the material used as a precursor, the presence of calcite stands out as the main peak, along with other phases such as kolvordskite or vaterite. The XRD spectra of ABA (Fig. 2 (b)) showed diverse phases with the presence of potassium as fairchildite or bütschilite. In the XRD of the silica sources RHA, SDE and BLA (Fig. 2 (c), (d) and (e)) the main peaks

Table 2
Chemical composition of the raw materials.

Oxides (% wt.)	BFS	ABA	RHA	SDE	BLA
SiO ₂	33.38	1.46	77.28	71.64	78.33
Al ₂ O ₃	10.94	0.45	0.37	8.51	4.96
Fe ₂ O ₃	2.00	0.48	0.14	1.27	5.79
MgO	6.52	2.91	0.62	0.53	0.97
CaO	41.32	26.69	1.39	0.68	2.36
Na ₂ O	0.26	0.24	1.02	1.76	0.11
K ₂ O	0.44	49.73	5.04	2.98	1.29
MnO	0.33	0.02	0.11	0.03	0.11
TiO ₂	0.46	0.05	0.01	0.20	1.11
P ₂ O ₅	0.02	4.46	1.10	0.16	0.66
LOI ^a	3.15	12.50	13.48	11.53	3.60
Trace elements (mg kg ⁻¹)	BFS	ABA	RHA	SDE	BLA
As	2	<2	21	8	2
Ba	923	44	36	919	119
Cd	<0.5	<0.5	<0.5	3	<0.5
Cr	21	46	25	62	86
Cu	7	154	14	20	60
Hg	<0.005	<0.005	0.057	<0.005	0.009
Mo	<2	2	3	6	<2
Ni	3	43	7	48	20
Pb	<5	<5	<5	18	<5
Sb	0.2	0.3	<0.2	2	<0.2
Se	<3	<3	<3	<3	<3
V	31	<5	<5	92	99
Zn	19	51	119	62	137

^a Loss on ignition at 750 °C.

observed were related to the presence of silica in its various forms of crystallization; quartz, cristobalite and tridymite.

3.2. Compliance leaching assessment of raw materials and AAMs

The compliance leaching test (EN 12457-4) is conducted to evaluate the potential release of trace elements from raw materials and crushed AAMs. The concentrations of eluates are compared with the European landfill Waste Acceptance Criteria (WAC) limit values for inert waste (Table 3).

The pH value of the raw materials is within the alkaline range, except for SDE. Conductivity values are particularly high in ABA because of the dissolution of the potassium carbonates and sulphates detected in the mineralogical characterization. RHA conductivity is also high, mainly due to the dissolution of sylvine (KCl), as reflected by the high concentrations of Cl⁻ in leachate. The leaching values of all elements in BFS are below the limits proposed by the European WAC (European Council, 2003) to be classified as inert waste. However, the rest of the materials surpass at least one of the limits considered. In the case of the alkali source ABA, the concentrations of Cr, Mo, Sb and SO₄²⁻ surpass these thresholds, while Cu, Se and V are close but remain below them. The alternative silica sources also do not meet the criteria to be classified as inert waste. Apart from the release of Cl⁻ and SO₄²⁻, particularly concerning is the release of As from RHA (10.705 mg kg⁻¹) which surpasses the inert and non-hazardous threshold of 2 mg kg⁻¹ (European Council, 2003). Release of Cd and Ni in SDE is slightly above the limits and is the highest of all materials, which may be promoted by the mildly acidic pH value of the leachate. SDE also exceeds the limits for V and F⁻. Lastly, the BLA surpasses the thresholds considered for V and SO₄²⁻.

Attending to the AAM leaching characteristics, the leachates pH values are strongly alkaline and are generally attributed to various factors such as the effectiveness of alkali activation or dosage of the activators (Sun et al., 2019). In this case, no significant differences in pH values and conductivity are perceived between the three activated materials. The trace elements release depicts that the three AAM samples are below all inert limit values. Trace elements and anions that previously had concentrations in the leachates of the raw materials above or near the limits are considerably decreased in the leachates of the AAMs.

Particularly, divalent cationic elements such as Cd, Ni, Pb and Zn are effectively immobilized with concentrations below or near the detection limit of the equipment, while the release of anions Cl⁻, F⁻ and SO₄²⁻ are also reduced compared to the secondary raw materials but remains close to the limits.

The mobility of oxyanionic elements As, Cr, Mo, Sb, Se and V, whose limits were surpassed by the secondary materials used as activators, are now reduced. The As is severely reduced, particularly in the sample AAM-RHA, that contains RHA as silica source. Cr released levels are also reduced considering that the three AAMs contain ABA as the alkali activator. However, a less significant effectiveness of immobilization is found in Mo, Se and V in the three samples, where released concentrations are below the limits but are not significantly reduced. In fact, the retention of elements that form oxyanions is generally poor in AAM (Keulen et al., 2018; Lancellotti et al., 2015). Hence, taking this into account and the results shown above, elements As, Cr, Mo, Sb, Se and V are considered critical when evaluating the leaching behavior of AAM and will be monitored in the subsequent leaching characterization.

3.3. Service life leaching assessment

As a monolithic construction material could be exposed to water for a long period of time, assessment of the dynamic (as a function of time) leaching behavior in the service stage of an AAM is crucial to estimate the potential of release of trace elements in the long-term. Therefore, the DSLT (CEN/TS 16637-2) is performed to the three monolithic AAM mixtures. The pH values and conductivity of the leachates as a function of time are depicted in Fig. 3(a) and (b), respectively.

pH values are initially maintained between 10.5 and 11, but a slight decrease is observed in the last stages of the test. Differences between the three AAMs are not significant, only in the long-term (>30 days) small variations appear, with the lowest values of pH in AAM-RHA. Conductivity, on the other hand, describes a significant decrease in the short-term stages, whilst in the last stages constant values are maintained. No significant differences are observed when comparing the conductivity over time of the three AAMs.

The trace element concentrations were represented as the cumulative release per exposed area of the monolithic samples, as a function of time (Fig. 4). The released concentrations are slightly higher in the AAM-RHA sample, except in the Sb. The most significant difference with the other two samples is found in the highest As release. Despite the difference in the As release, the tendencies as a function of time are the same for the three AAM. Considering the rest of the trace elements Cr, Mo, Sb, Se and V, the release curves over time are almost similar between samples, and consist in a sharp increase of concentrations in the short time stages (0.25–4 days) followed by a moderate increment in release for the long-term stages (from 16 until 64 days).

Total cumulative release values at 64 days from the DSLT are compared to the limit values of Soil Quality Decree (SQD) from the Netherlands (Table 4). SQD values are selected because of the absence of harmonized limit values for monolithic leaching assessment in the EU, and it is employed in other studies that address leaching behavior of construction materials and AAM (Keulen et al., 2018; Sun et al., 2022; Taha et al., 2018).

Although the As release from AAM-RHA is significantly higher, the concentrations are far below the thresholds of the SQD. The release of the other elements is also significantly lower compared to the limit values, and no significant differences are found between samples, indicating a relatively low mobility of oxyanionic elements in the three monolithic AAM.

The information about the release mechanisms from AAM is important to understand the potential leaching behavior of the material in the long-term. In this case, the mechanisms are determined in a stepwise procedure following the calculations in CEN/TS 16637-2 (European Committee for Standardization, 2014). A flowchart that includes the equations and steps followed to determine the release

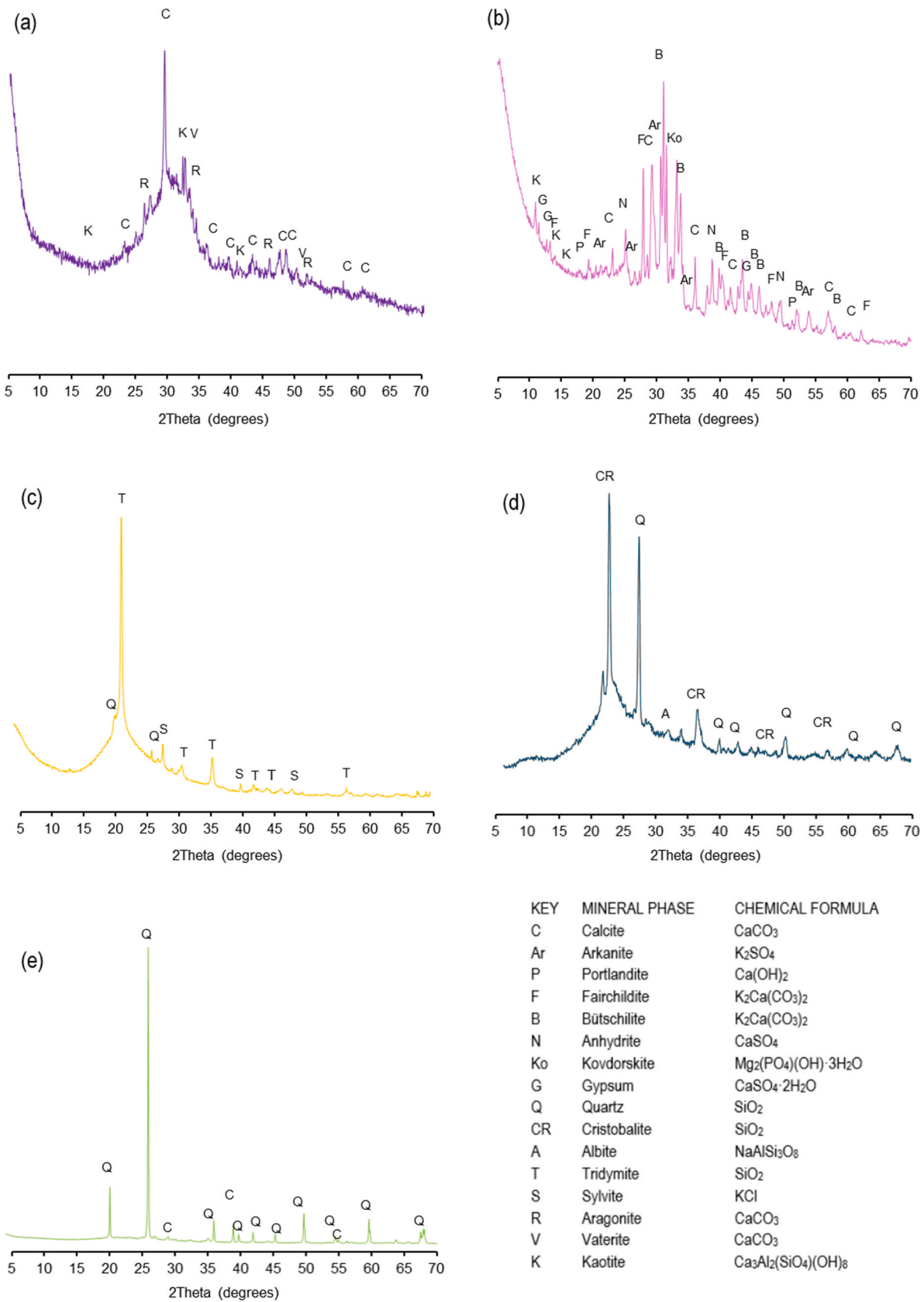


Fig. 2. XRD pattern of (a) BFS; (b) ABA; (c) RHA; (d) SDE and (e) BLA.

Table 3

pH values, conductivity, and elements concentration in leachates of raw materials and AAM from compliance leaching test (EN 12457-4) in comparison with WAC inert waste limits (bolded values surpass the limit).

Elements (mg kg ⁻¹)	BFS	ABA	RHA	SDE	BLA	AAM-RHA	AAM-SDE	AAM-BLA	Limit values
pH	12.11 ± 0.014	10.64 ± 0.007	9.93 ± 0.014	4.76 ± 0.035	9.01 ± 0.007	11.92 ± 0.028	12.01 ± 0.078	11.85 ± 0.071	–
Conductivity (µS cm ⁻¹)	3,055 ± 35.36	32,850 ± 636.40	6,200 ± 141.42	921.5 ± 54.45	1,004 ± 2.12	3,655 ± 332.34	3,585 ± 770.75	3,100 ± 424.26	–
As	0.001 ± 0.0002	0.092 ± 0.0019	10.705 ± 0.828	0.151 ± 0.007	0.150 ± 0.016	0.022 ± 0.004	0.023 ± 0.009	0.010 ± 0.003	0.5
Ba	7.628 ± 0.130	0.325 ± 0.009	0.001 ± 0.0001	0.807 ± 0.008	0.369 ± 0.046	0.101 ± 0.001	0.089 ± 0.005	0.124 ± 0.0003	20
Cd	<DL	0.001 ± 0.0001	0.0004 ± 0.00001	0.051 ± 0.003	<DL	<DL	<DL	<DL	0.04
Cr	0.068 ± 0.001	5.404 ± 0.128	0.131 ± 0.010	0.415 ± 0.011	0.011 ± 0.001	0.020 ± 0.003	0.013 ± 0.004	0.014 ± 0.006	0.5
Cu	0.004 ± 0.0004	1.345 ± 0.018	0.002 ± 0.002	0.237 ± 0.004	0.005 ± 0.001	0.002 ± 0.0003	0.015 ± 0.0030	0.007 ± 0.0030	2
Hg	0.002 ± 0.0003	0.001 ± 0.0003	0.003 ± 0.0001	0.001 ± 0.0002	0.0004 ± 0.00002	0.0004 ± 0.0001	0.0003 ± 0.0001	0.0003 ± 0.0001	0.01
Mo	0.063 ± 0.0004	1.106 ± 0.004	0.393 ± 0.033	0.398 ± 0.016	0.169 ± 0.047	0.147 ± 0.024	0.199 ± 0.111	0.126 ± 0.041	0.5
Ni	0.001 ± 0.00001	0.309 ± 0.0001	0.001 ± 0.0003	0.632 ± 0.023	0.002 ± 0.0002	0.001 ± 0.0003	0.010 ± 0.003	0.006 ± 0.002	0.4
Pb	0.0002 ± 0.00004	0.009 ± 0.0002	0.001 ± 0.0004	0.008 ± 0.0001	<DL	<DL	<DL	<DL	0.5
Sb	0.0003 ± 0.00002	0.096 ± 0.001	0.004 ± 0.0003	0.039 ± 0.0008	0.002 ± 0.0002	0.004 ± 0.0003	0.006 ± 0.001	0.003 ± 0.001	0.06
Se	0.015 ± 0.0001	0.090 ± 0.002	0.056 ± 0.002	0.029 ± 0.0005	0.050 ± 0.002	0.075 ± 0.016	0.056 ± 0.005	0.072 ± 0.019	0.1
V	0.007 ± 0.00004	1.068 ± 0.005	0.070 ± 0.006	3.453 ± 0.122	1.854 ± 0.226	0.424 ± 0.064	0.446 ± 0.136	0.405 ± 0.151	1.8 ^a
Zn	0.007 ± 0.004	0.075 ± 0.019	0.028 ± 0.006	1.469 ± 0.107	<DL	<DL	<DL	<DL	4
Cl ⁻	434.67 ± 0.431	647.68 ± 4.96	11,585.23 ± 413.37	48.99 ± 4.55	392.69 ± 0.417	329.82 ± 57.35	145.94 ± 21.21	165.64 ± 46.84	800
F ⁻	3.85 ± 0.049	2.49 ± 0.106	3.57 ± 0.035	67.73 ± 9.69	2.98 ± 0.09	4.33 ± 0.70	8.08 ± 1.89	4.46 ± 1.40	10
SO ₄ ²⁻	50.26 ± 2.58	4,477.64 ± 118.46	4,627.55 ± 183.49	654.43 ± 70.27	3,204.47 ± 7.34	904.71 ± 156.96	699.89 ± 135.82	907.53 ± 266.50	1000

<DL – Under detection limit of the apparatus.

^a Adopted from Soil Quality Decree limit values for unmolded materials (Soil Quality Decree, 2007).

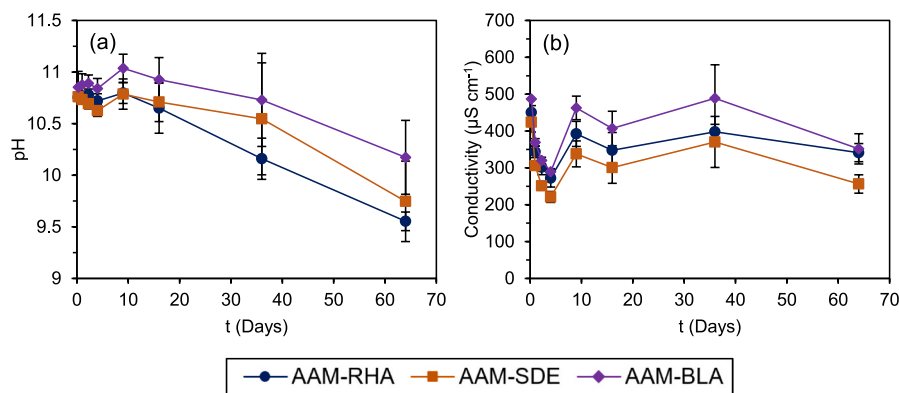


Fig. 3. Evolution of (a) pH values and (b) conductivity in DSLT (CEN/TS 16637-2) of monolithic AAM.

mechanisms is supplied in Fig. S1. The controlling release mechanisms can be divided in primary mechanisms (diffusion and dissolution) and secondary mechanisms (surface wash-off and depletion) and are derived from the experimental data in DSLT, and the determined mechanisms for monolithic AAM are shown in Table 5.

It is found that release mechanisms identified for all the trace elements are the same for the three AAM samples. The prevalent primary release mechanism is diffusion, determined for Cr, Sb, Se and V, while for As and Mo remains unidentified. In the case of Se and V, an initial surface wash-off is also determined preceding the diffusion primary

mechanism. In the case of As, the secondary mechanism surface wash-off is determined for all the mixtures. For Mo, despite the released concentrations as a function of time are quite similar to the elements identified as diffusion-controlled release, the primary release mechanism remains unidentified and surface wash-off is identified as the secondary release mechanism.

3.4. End-of-life leaching assessment

Prior to reuse, the demolished granular material can be left in

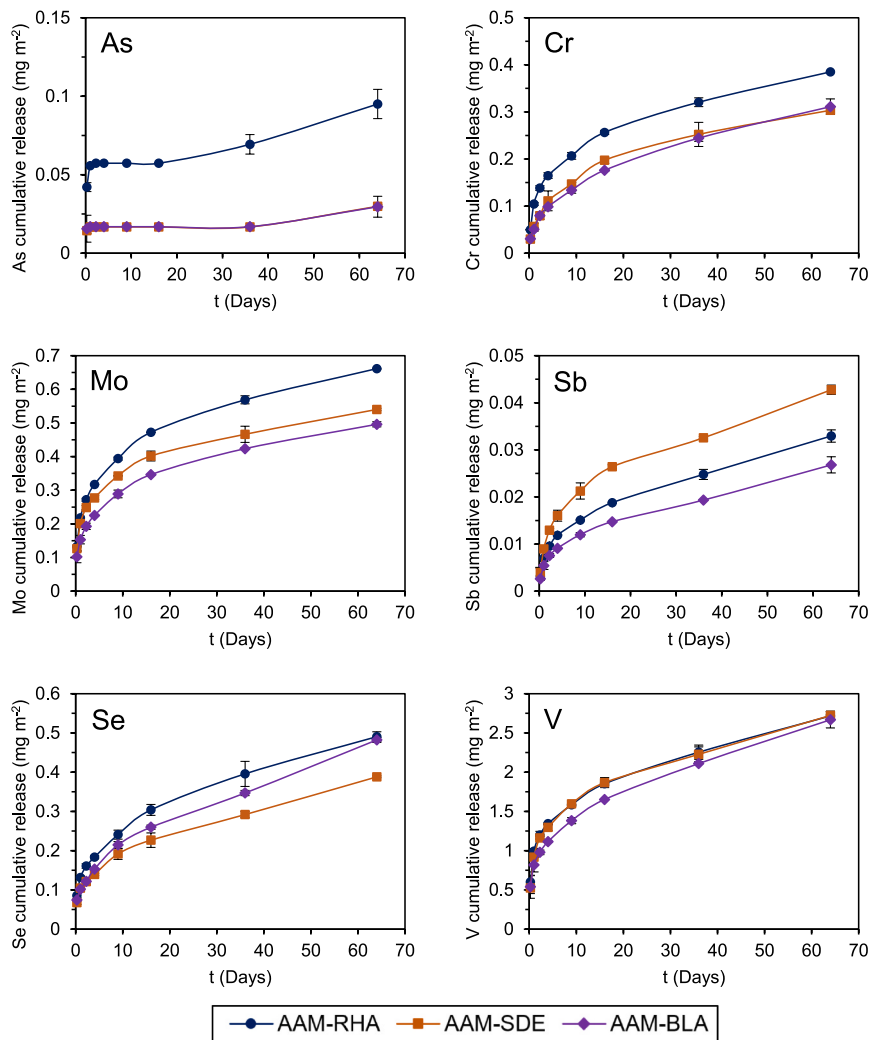


Fig. 4. Cumulative release (mg m^{-2}) of trace elements in DSLT (CEN/TS 16637-2) of monolithic AAM.

Table 4

Cumulative release values at 64 days of AAM and Dutch soil quality decree limit values for unrestricted use of monolithic construction materials.

Elements (mg m^{-2})	AAM-RHA	AAM-SDE	AAM-BLA	SQD limits
As	0.095	0.029	0.029	260
Cr	0.38	0.30	0.31	120
Mo	0.66	0.54	0.50	144
Sb	0.033	0.042	0.027	8.7
Se	0.49	0.39	0.48	4.8
V	2.72	2.72	2.67	320

controlled or uncontrolled leaching scenarios where water may infiltrate through the solid due to exposure to rainwater or seepage, opening the possibility of release of hazardous substances. Therefore, the end-of-life leaching characteristics of a waste-based material should be assessed through a test that resembles this scenario (Engelsen et al., 2021). The up-flow percolation leaching test (CEN/TS 16637-3) is performed to check the influence of water passing through the granular AAM (progressively increased L/S ratio).

The pH values (Fig. 5 (a)) initially are strongly alkaline ($\text{pH} > 12$), which is caused by the dissolution of alkaline species and soluble salts from the crushed AAM, and are slightly decreased as L/S increases. On the contrary, conductivity (Fig. 5 (b)) is severely decreased due to dissolution of excess of alkaline constituents of the AAM, also explaining

the parallel decrease in the pH values of the eluates. pH and conductivity values of AAM-BLA in the first extracted eluates are slightly higher than the AAM-SDE and AAM-RHA. Despite this, values are found to be very similar for the three mixtures as L/S ratio is increased, and significant differences are not found between samples.

The concentrations in the percolation test leachates were represented as the cumulative trace element release per kilogram of dry matter versus the L/S ratio (Fig. 6).

The tendencies described for all samples consist in a significant rise in the release of elements in the shortest L/S ratios, followed by a decrease in the final stages which is, however, different depending on the element considered; in As, Mo and Se is more pronounced than in Cr and Sb. The As release is the highest in the AAM-RHA, but released concentrations of AAM-SDE remain close, despite not containing RHA. Indeed, except for Cr, the concentrations of Mo, Sb, Se and V are the highest in the AAM-SDE sample.

The release mechanisms of the granular AAM were determined from the experimental results of the up-flow percolation leaching test according to the procedures in CEN/TS 16637-3 (European Committee for Standardization, 2016). The mechanism for all the samples and all the trace elements was found to be “apparent depletion”. This expression stands for the depletion trend of the trace elements, which is in line with the observed cumulative release, but concentrations are not sufficiently low (i.e., close to detection limits of the apparatus) to ascertain that release of substances will be completely depleted in the long-term.

Table 5
Release mechanisms of trace elements for monolithic AAM mixtures according to procedure in CEN/TS 16637-2.

Release Mechanisms	AAM-RHA						AAM-SDE						AAM-BLA					
	As	Cr	Mo	Sb	Se	V	As	Cr	Mo	Sb	Se	V	As	Cr	Mo	Sb	Se	V
M1 - Overall low concentrations	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M1.1 - Surface wash-off following M1	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M2 - Diffusion	-	✓	-	✓	-	-	-	✓	-	✓	-	-	-	✓	-	✓	-	-
M2.1 - Surface wash-off preceding M2	-	-	-	-	✓	✓	-	-	-	-	✓	✓	-	-	-	-	✓	✓
M2.2 - M2 with depletion	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M2.3 - M2.1 and M2.2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M3 - Dissolution	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M4 - Unidentified mechanism	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M4.1 - Surface wash-off preceding M4	✓	-	✓	-	-	-	✓	-	✓	-	-	-	✓	-	✓	-	-	-
M4.2 - Depletion following M4	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M4.3 - M4.1 and M4.2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Determined release mechanism	M4.1	M2	M4.1	M2	M2.1	M2.1	M4.1	M2	M4.1	M2	M2.1	M2.1	M4.1	M2	M4.1	M2	M2.1	M2.1

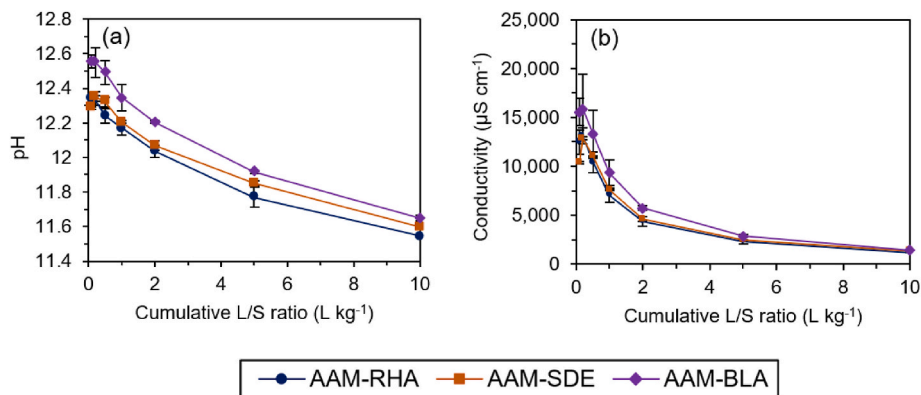


Fig. 5. (a) pH values and (b) conductivity in up-flow percolation leaching test of granular AAM.

The released cumulative concentrations in the percolation test are compared with the limit values for the unrestricted use of unmolded building materials from the Dutch SQD (Table 6). This criterion is specifically set for the evaluation of granular building materials at a L/S ratio of 10 in a percolation leaching test. The released values from the three AAM mixtures are far from the limits of all the oxyanionic elements. Therefore, the material can be considered environmentally acceptable to be reused in other applications (e.g., as recycled aggregate).

4. Discussion

This research explored the leaching characteristics of AAM with alternative activators derived from agro-industrial waste that contain trace elements of potential concern when released to the environment.

The evaluation of the leaching potential of the AAM through the compliance leaching test showed reduced concentrations of trace elements, below the limit values of the WAC, demonstrating the effective inertization of the elements that mobilized in the waste-derived activators when they are incorporated to the AAM. Retention or release of trace elements in AAM is dependent on the type of mix design and the chemical content of the starting materials, as noticed by Keulen et al. when varying the quantity of pulverized coal fly ash (FA) in FA-BFS hybrid AAM mixtures (Keulen et al., 2018). In fact, other studies

showed a several increase in mobilization of As when activating municipal solid waste incineration (MSWI) ashes with commercial NaOH solutions (Álvarez-Ayuso et al., 2008; Maldonado-alameda et al., 2021). On the other hand, when BFS is employed as precursor its high calcium content promotes the As, Cr and Cd fixation in the AAM (Lancellotti et al., 2015). For instance, in this work, the main source of As is the RHA as silica-based activator, which is greatly immobilized when incorporated in the AAM-RHA sample. Therefore, the synergistic use of BFS as precursors and agro-industrial wastes as alternative activators immobilizes the trace elements contained in the latest.

In relation to the service life assessment of AAM, same range of pH values and slight decrease in the latest stage of the test is also observed by Keulen et al. (2018) in AAM mixtures with pulverized coal fly ash and BFS as precursors and NaOH as activator solution, and release over time of oxyanionic trace elements is in agreement with the tendencies found for AAMs with BFS as main precursor and NaOH and waterglass as activators (Sun et al., 2022). The high concentrations of As in AAM-RHA compared the AAM-SDE and AAM-BLA samples are attributed to the incorporation of RHA in the sample, and may be caused by the dissolution of soluble species (e.g. dissolution of mineral phase where As was bonded) (Engelsen et al., 2021) or residual, non-reacted fractions of RHA. Still, concentrations of As and the other trace elements characterized are significantly low, as they are far from the limits considered. Indeed, the cumulative concentrations at 64 days are within the range of

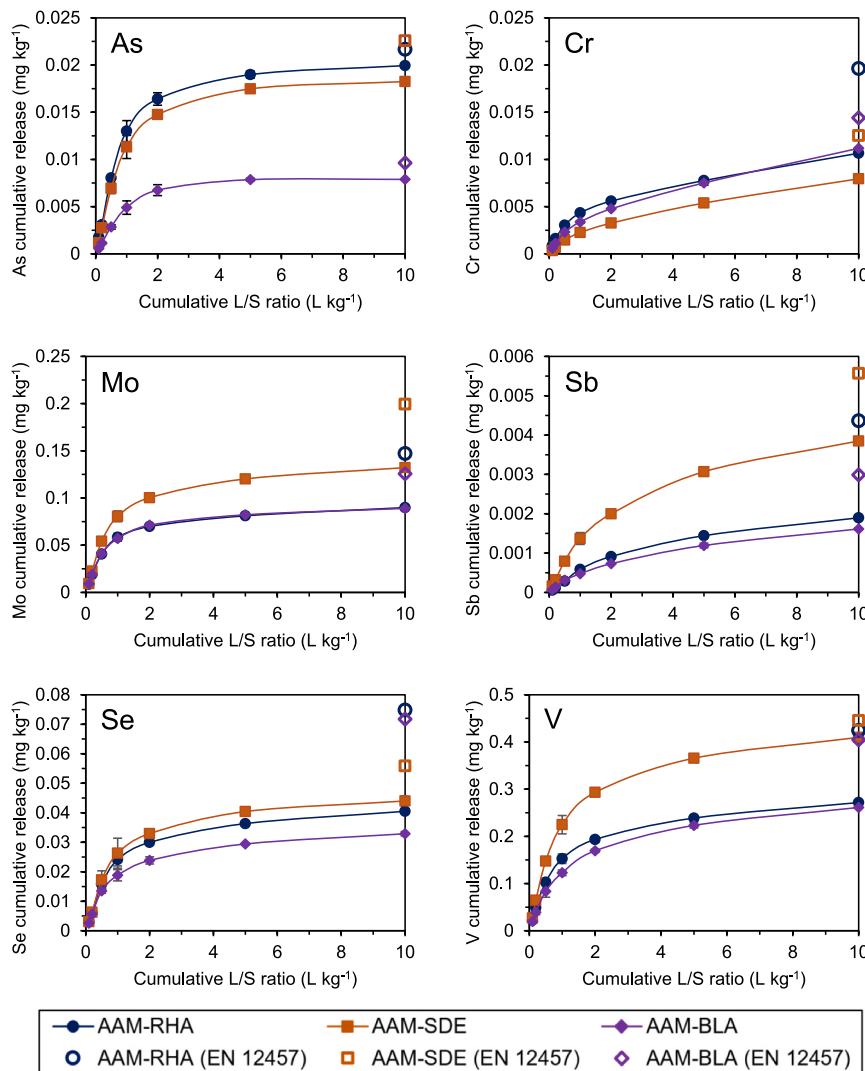


Fig. 6. Cumulative release (mg kg⁻¹) of trace elements in percolation test and compliance leaching test values of granular AAM.

Table 6

Cumulative release values at cumulative L/S = 10 of AAM in up-flow percolation test and Dutch soil quality decree limit values for unrestricted use of granular materials.

Elements (mg kg ⁻¹)	AAM-RHA	AAM-SDE	AAM-BLA	SQD limits
As	0.020	0.018	0.008	0.9
Cr	0.011	0.008	0.011	0.63
Mo	0.090	0.132	0.089	1
Sb	0.002	0.004	0.002	0.16
Se	0.040	0.044	0.033	0.15
V	0.272	0.410	0.261	1.8

values determined for ordinary Portland cement-based concretes (Hartwich and Vollpracht, 2017; Sun and Vollpracht, 2020) and below the determined concentrations of AAMs made of commercial activators (Maldonado-Alameda et al., 2022; Sun et al., 2022).

Regarding the monolithic leaching mechanisms, impossibility of the determination of the controlling release mechanism could be due to changes in pH or low concentrations determined in the eluates or a combination of both (Sun and Vollpracht, 2020). Particularly for Mo, although released concentrations over time are similar to elements which release mechanism is established as diffusion, the indetermina-tion of the primary controlling mechanism is caused by the sensitivity of

the parameters, particularly of the square root of the mean squared error ($\sqrt{\text{MSE}}$) parameter as the only condition to be complied for diffusion identification, that should be lower than 0.40 (European Committee for Standardization, 2014). Values determined for the three samples are sensibly higher and therefore criterion is not met for the three mixtures. Indeed, the procedure will not always be able to effectively determine the release mechanism (Hartwich and Vollpracht, 2017; Sun and Vollpracht, 2020).

In relation to the granular leaching behavior, comparing the concentrations obtained in the compliance leaching test and the cumulative release at L/S = 10 L/kg in the percolation test, it is observed that concentrations determined in the compliance tests are slightly higher but remain relatively close to release in percolation conditions. This relation between batch and column tests is also found by Giels et al. (2019) for oxyanionic elements in AAM based on metallurgical slags. Thus, the compliance leaching test can be considered a good conservative approximation to the results of a percolation test when characterizing the trace element release of a granular AAM.

On the other hand, it should be remarked that no relationships between granular and monolithic leaching can be established. The highest release of trace elements from AAM-SDE granular samples in the percolation test (Fig. 6) contrasts with the highest released concentrations determined from AAM-RHA monolithic samples in the DSLT (Fig. 4). Also, the diffusion as prevalent release mechanism in the

monolithic form differs from the apparent depletion determined for all the trace elements in the granular material. This fact emphasizes the necessity of a comprehensive leaching characterization of the material depending on their intended use scenario, and not only on legal requirements (Engelsen et al., 2021).

Although apparent depletion was determined as the only mechanism in granular AAM and concentrations are not sufficiently low to determine full depletion of the trace elements release, it should be remarked that the release is not very prominent and the three AAMs are assumed to retain contaminants very efficiently when evaluating their leaching in the end-of-life (granular) scenario.

Summarizing, no significant differences were found in the leaching behavior of AAMs when modifying the silica-based alternative activator (RHA, SDE and BLA), and, according to the comparison with literature, these AAMs activated with agro-industrial waste show similar or better leaching characteristics of trace elements of potential concern than AAMs made of commercial activators, and they can be considered environmentally compatible in their monolithic (service life) and granular (end-of-life) form since they comply with the SQD limit values.

5. Conclusions

The BFS based AAM activated with agro-industrial waste were assessed through a combination of horizontal leaching tests. The alternative activators, ABA (as alkali source) and RHA, SDE and BLA (as silica rich source), surpassed the Inert Waste Acceptance Criteria limits for some trace elements, and were inertized by the activation process. AAM developed complied with the Dutch Soil Quality Decree limits for monolithic and granular construction materials, which assures their environmental acceptability throughout their life cycle. In addition, no significant influence of the type of silica source on the leaching behavior was observed, demonstrating the environmental compatibility of the substitution of commercial silicate with these secondary silica sources in AAMs. Long-term leaching behavior of monolithic AAM samples from the Dynamic surface leaching test results demonstrated that the predominant release mechanism for the Cr, Sb, Se and V is the diffusion and for As and Mo is surface wash-off, whilst in the granular AAM samples from the Up-flow percolation leaching test results is determined for the trace elements the apparent depletion. The non-correlation of the results suggests the need for an entire life cycle leaching assessment of the waste-derived AAMs. Based on the results obtained, future research should focus on the geochemical modelling of trace elements in AAM to determine the leaching controlling factors, in order to optimize the design of the waste-derived alkali activated formulations.

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CRediT authorship contribution statement

Iván Salas: Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. **Eva Cifrián:** Writing – review & editing, Supervision, Conceptualization. **Lourdes Soriano:** Writing – review & editing, Resources, Investigation. **José Monzó:** Funding acquisition. **Ana Andrés:** Writing – original draft, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2024.122265>.

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