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Recycling belite cement clinker from post-demolition autoclaved aerated concrete – assessing a new process

P. Stemmermann^a, R. Volk^{b,*}, J.J. Steins^b, G. Beuchle^a

^a Karlsruhe Institute of Technology (KIT), Institute for Technical Chemistry (ITC), Postfach 3640, 76021, Karlsruhe, Germany

^b Karlsruhe Institute of Technology (KIT), Department of Economics and Management, Institute for Industrial Production (IIP), Hertzstr. 16, 76187, Karlsruhe, Germany

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ABSTRACT

Increasing post-demolition autoclaved aerated concrete (pd-AAC) waste is mainly landfilled due to its physical properties and lacking recycling processes. A promising technology is the production of recycled belite cement clinker, which can partially substitute Portland cement clinker. This paper presents experimental data of recycled belite cement clinker production from pd-AAC that has been successfully demonstrated on technology readiness level 4–5 and its associated lifecycle assessment. Different supply chains for pd-AAC and energy are examined. The closed-loop pd-AAC recycling via the belite route that aims for Portland cement clinker substitution shows significant potential savings in environmental impacts. These savings could reach 0.77 kg CO₂-Eq/kg pd-AAC compared to the status quo (landfilling) by using renewable electricity, and 0.34 kg CO₂-Eq/kg pd-AAC by using natural gas. The gained reduction of around 13.5 % is significant considering that it is the result of substituting only 15.5 % of the overall input material.

Spotlights

- As of today, mineral waste fines from autoclaved aerated concrete are landfilled; a new technology recycles it to cement clinker that substitutes cement in processing of new autoclaved aerated concrete
- Various environmental aspects are improved, especially with regard to CO₂-equivalent savings and landfill capacity
- Substitution of Portland cement shows significant potential savings of up to 0.77 kg CO₂-equivalents per kg input
- However, when using the German electricity mix, there is some worsening of other environmental impacts
- Future research should work towards a pilot plant, system analysis and network design for optimal circularity

1. Introduction

Autoclaved aerated concrete (AAC) is a popular mineral lightweight construction material (Steins et al. 2022). AAC is highly porous, with a total pore volume of 65 to 90 % (Schober 2011). The bridges between mm-sized air pores consist of the calcium silicate hydrate tobermorite

and smaller quantities of other minerals, especially ones containing sulfate and unreacted raw materials, especially quartz. The raw materials include ground quartz, ordinary Portland cement (OPC), quicklime, gypsum, and aluminum powder. After mixing with warm water the resulting slurry is poured in forms. Slow hardening generates a cake that rises by released hydrogen as propellant gas. During the reaction, the volume of the mixture increases 2 to 3-fold, leading to a low density. Finally, the cake is hardened in an autoclave under hydrothermal conditions. Thus, AAC has excellent thermal insulation properties and can be easily assembled on the building site. Typically, in Europe, AAC is produced in plants with a production capacity of between 110,000 and 275,000 m³/a (UBA 2019; Harder 2009). Considerably used since around the 1950s, a rising amount of post-demolition AAC (pd-AAC) is expected to return from the building stock soon (Steins et al. 2021, 2022). Pd-AAC is ‘generated’ locally as a separate or mixed demolition fraction. In Germany it is jointly collected with other gypsum-containing demolition waste and is usually landfilled (Kreft 2016; UBA 2019).

Recent studies address new open-loop recycling options for pd-AAC, e.g. in lightweight aggregate concrete production (Aycil et al. 2016; Gyurkó et al. 2019), light mortar production (Aycil et al. 2016), floor

Abbreviations: AAC, autoclaved aerated concrete; C₂S, belite (2CaO.SiO₂); C₃S, alite (3CaO.SiO₂); OPC, ordinary Portland cement; PCC, Portland cement clinker; pd-AAC, post-demolition AAC; RC-BCC, recycled belite cement clinker.

* Corresponding author.

E-mail address: rebekka.volk@kit.edu (R. Volk).

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screed production (Bergmans et al. 2016), filler or supplementary material in concrete (Gyurkó et al. 2019), cement clinker production (Schoon et al. 2013), and concrete (deprived of the fine fraction) for specific applications and shuttering blocks (Gyurkó et al. 2019). Volk et al. (2022) and Volk et al. (2023) compare the recycling options in a life cycle assessment (LCA). However, these options are not closed-loop and might be limited with growing mass flows.

Moreover, pd-AAC qualities differ strongly from pure to heavily mixed with wood, glass, metal, screw anchors, gypsum/plastering, wallpaper, color, or other coatings. Average compositions of variously collected pd-AAC are shown in Table A-1 (in supporting material A). Additionally, pd-AAC has a very heterogeneous fragment size. Therefore, prior to any recycling, preprocessing by various crushing and sorting steps is necessary. Most recycling options focus on granules above 2 mm grain size. However, this material class is a minor fraction of the preprocessing with approximately ¼ share of the input mass. The main fraction is fines (ca. ¾),¹ which is hardly recycled today. Besides, the sulfate content increases significantly in the fines fraction.

Due to the use of raw materials and the reaction with water, AAC is similar in composition to a hardened Portland cement paste but deficient in CaO. Therefore, it is appealing to use pd-AAC fractions in a chemical recycling process to produce Portland cement if there is no better physical option for recycling. However, to use pd-AAC as a raw meal substitute, its calcium oxide content must be increased by adding limestone (CaCO₃). The subsequent clinker burning includes calcination of limestone as an essential step, which requires much energy and releases CO₂.

To reduce the specific CO₂ emissions and to broaden the range of suitable raw materials for clinker production, high-belite (2CaO.SiO₂ or C₂S) cement can be produced (Kotsay et al. 2019). However, production is only economically viable at temperatures above 1200 °C in high-capacity plants. Therefore, a new type of belite cement clinker, recycled belite cement clinker (RC-BCC), has recently been developed at KIT. Its chemistry complies with the European cement standard (DIN EN 197-1) for Portland cement clinker (PCC), with a belite content of up to 80 wt%. Special conditions in the kiln allow clinkering at a much lower temperature (RC-BCC 1000 °C vs. PCC 1450 °C) (Ullrich et al. 2021; Beuchle et al. 2013), which lowers energy consumption and enables electric heating with available technologies instead of combustion. Environmentally harmful and cement-damaging impurities are fixed by incorporation into non-soluble minerals, which allows for low-quality secondary raw materials. The slower hydration kinetics of RC-BCC (Chatterjee 1996; Chen et al. 2017) is compensated by formulating mixed types of cement from alite-rich (3CaO.SiO₂ or C₃S in cement notation) PCC, RC-BCC, or other main constituents according to European standards.

The RC-BCC technology reduces the specific consumption of limestone by about 10 % and substitutes limestone with CO₂-free secondary materials such as pd-AAC. Both features reduce the specific CO₂ emission and energy consumption of RC-BCC relative to PCC. In addition, natural resources are saved, and waste is reduced. Furthermore, the lower process temperature simplifies the kiln design. Finally, the technology enables easy separation of concentrated CO₂ if process heat comes from oxyfuel combustion or electricity. Both the use and storage of highly concentrated CO₂ are economically favourable.

Therefore, this study aims to describe the technical process of RC-BCC production, including chemical reactions, material balances, energy consumption, and CO₂ emissions (Section 2). Moreover, a full LCA is conducted to assess the environmental potential of RC-BCC production from pd-AAC and the results are compared to status quo (land-filling) (Section 3). Finally, the results are discussed (Section 4) and

¹ Expert interview within research project REPOST with Xella Technologie- und Forschungsgesellschaft mbH. However, the shares can vary due to water content, the used mill (Krampitz et al. 2022) and other factors.

concluded (Section 5).

2. Recycled belite cement-clinker (RC-BCC): process description and assessment basics

2.1. General process design

Processing of RC-BCC (Fig. 1) starts with the pre-treatment of the raw materials. The main components are crushed, graded, purified, and, if necessary, pre-dried. Each component is ground until 50 % of the particles are smaller than 20 µm ($d_{50} = 20 \mu\text{m}$) with $d_{95} < 100 \mu\text{m}$. The grinding time depends on the components and is usually 5 to 15 min (Cement equipment organisation 2018). The resulting grain size distribution is checked by static light scattering. The powdery products are mixed into a raw meal with a CaO/SiO₂ molar ratio of two. The mixture is stored in a silo and analyzed. Depending on its composition, corrective limestone as well as mineralizer is added. The mixture reacts to RC-BCC in a rotary kiln at 1000 °C in a slightly oxidizing CO₂ atmosphere and is cooled in a clinker cooler. Finally, the clinker is ground in a ball mill to achieve a typical cement fineness.

2.2. Firing conditions

Three heating technologies have been considered (Table 1). The simplest variant uses the combustion of natural gas in air, which results in large amounts of off-gas with low CO₂ concentration. The second variant, the oxyfuel technology, combusts natural gas in an oxygen (-rich) atmosphere. In the third variant, the rotary kiln is electrically heated. However, heating technologies differ in their level of technological readiness.

2.3. Combustion and process gasses

Depending on the firing technology, the off-gas stream's composition, quantity, and energy content vary. The CO₂ concentration in the reactor strongly influences the reaction kinetics and, thus, the plant output. The use of secondary raw materials may induce impurities such as CO, SO₂/SO₃, HCl, or trace elements, which affect the quality of CO₂ for later use or storage. Incomplete oxidation is prevented for safety reasons by a slight oversupply of oxygen.² The process heat is used as far as possible to preheat the raw meal. In atmospheric combustion, the combustion air is preheated in the clinker cooler. In oxyfuel or electric heating, clinker cooling is done by heat-exchanged concentrated CO₂ of more than 90 % purity. The concentrated CO₂ can be used, e.g. in on-site carbonation of waste concrete (Vanderzee and Zeman 2018), or stored.

2.4. RC-BCC from pd-AAC and limestone: mixing ratio of the starting materials

Three pd-AAC samples (SM(D1), SM(D2), and SM(D3)) and one sample of pure production waste (SM(P)) were provided by a mineral waste processing company and an AAC producer. Details are described in (Ullrich et al. 2021). X-ray fluorescence analysis (XRF) is used to measure the chemical composition of the samples, which contain typically 43 to 56 wt% of SiO₂ and 25 to 31 wt% of CaO. Oxide contents are given in Table A-1 in supporting information A.

Bulk quantitative phase analyses of all samples were performed mixed with an internal standard (20 wt% $\alpha\text{-Al}_2\text{O}_3$, Alfa Aesar 99.95 %) using the Rietveld method following the fundamental parameters approach implemented in TOPAS V6 (Bruker-AXS) to determine crystalline and X-ray amorphous contents. A detailed description of the refinement strategy is given in Ullrich et al. (2022).

RC-BCC samples were synthesized from the four pd-AAC samples

² In scenario 1: oversupply of air.

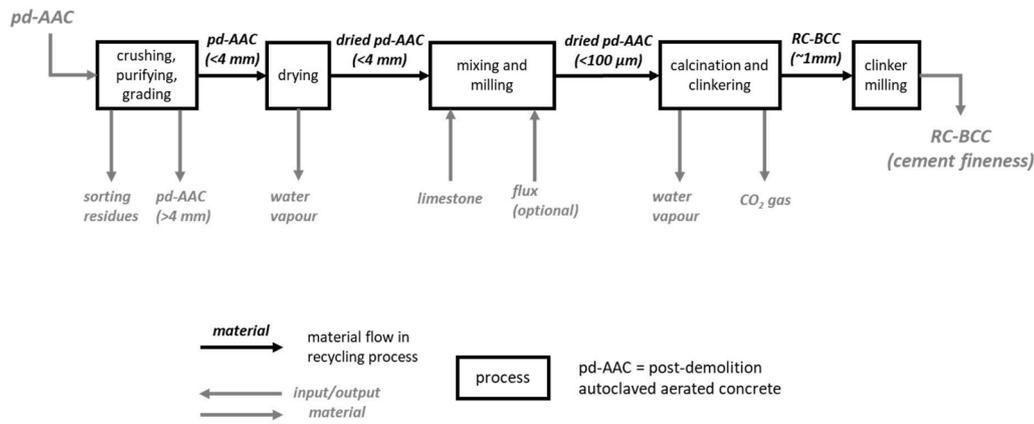


Fig. 1. RC-BCC production flow sheet.

Table 1
Overview of assessed process variants.

	Gas	Oxyfuel	Electric
Firing conditions	Combustion of natural gas	Combustion of natural gas	Electricity
Atmosphere in the rotary kiln	N ₂ +CO ₂ +5 % Oxygen	CO ₂ + 5 % Oxygen	CO ₂ + 5 % Oxygen
Technology readiness level (TRL)	Established standard (TRL = 9)	Not established for this temperature range (TRL = 5)	Not established for larger plant sizes (TRL = 4)
Addition of oxygen	No	Yes	Yes
Clinker cooling	Established standard (TRL = 9)	Yes, via heat exchange of CO ₂ (TRL 7)	Yes, via heat exchange of CO ₂ (TRL 5)

with respective limestone additions in a laboratory rotary kiln (Ullrich et al. 2021). Pure CaCO₃ was used in the experiments.

Depending on each sample's CaO and SiO₂ content, the addition of CaCO₃ is calculated to achieve a raw meal with a molar ratio of CaO to SiO₂ of two – or in cement notation (C = CaO; S=SiO₂): C/S = 2 see Table A-2 (supporting information A). Finally, a small addition of a mineralizer, typically 1 to 2 wt% of either Na₂CO₃ or CaCl₂ (Garbev et al. 2022; Ullrich et al. 2022), is added to deal with contaminations and increase the kinetics.

Energy and mass balances were calculated based on thermodynamic equilibrium calculations with the database system Factsage 8.2 (Bale et al. 2016). However, the available data sets for the respective systems do not allow for incorporating minor and trace elements in solid solutions. Thus, for stable results, the composition of pd-AAC was approximated with a data set for a simplified calculated average pd-AAC composition (SC-pd-AAC). It was derived from mean values for phase contents according to the ranges given in the safety sheet “YTONG autoclaved aerated concrete” (Xella 2017) and considering an additional 3% moisture in pd-AAC (Table A-1, A).

2.5. Chemical reactions and mineralogy of RC-BCC processing

The clinker raw meal is processed in a rotary kiln. During heating up to about 700 °C, it gradually expels the chemically bound water from mineral phases. After the loss of approx. 3% free moisture, these are in particular ~2 wt% crystal water from gypsum (150 °C) and 3.5 wt% from tobermorite (main weight loss up to 200 °C, then gradually). The total weight loss sums up to approx. 9 wt%.

At about 600 °C, calcium silicates from dehydrated pd-AAC and limestone begin to form and release CO₂. The availability of SiO₂ and CaO determines the reaction rate which can be enormously accelerated by adding mineralizer, e.g. CaCl₂ or Na₂CO₃. For combustion in air, the

temperature of limestone decomposition is reached at approximately 650–800 °C, depending on the limestone's crystallinity. Large amounts of free CaO are formed, slowly reacting with the other components and forming the desired clinker phases.

The decomposition temperature of limestone increases by about 100 °C, accompanied by the immediate formation of calcium silicates, if clinkering of RC-BCC is performed in a concentrated CO₂ atmosphere (electric heating, oxyfuel). Clinkering is finished at about 1000 °C. The clinker grains consist predominantly of belite, an x-ray amorphous mixture of highly disordered nanocrystals, and a vitreous fraction. The x-ray amorphous mixture's chemical composition corresponds to the one of the bulk samples. Sulfate is fixed in a sulfate-rich calcium silicate or sodium sulfate, depending on the used mineralizer. Without the addition of mineralizer, the sulfate-rich calcium silicate ternesite crystallizes. If CaCl₂ is used as mineralizer, chloro-ellestadite will form instead. Besides chlorine and sulfate, ellestadite can incorporate other potentially environmentally harmful or cement-damaging impurities in its crystal structure. It is insoluble under normal cementing conditions. When Na₂CO₃ is used as a mineralizer, Na₂SO₄ is formed. Na₂SO₄ is highly soluble in water and can be easily washed out. Table 2 compares the measured phase contents of RC-BCC with contents calculated with Factsage 8.2 (Bale et al. 2016) for the simplified calculated average pd-AAC composition SC-pd-AAC / 2 wt% CaCl₂. Since some of the phases present in the experimental data, e.g. ellestadite, do not appear in the published databases, the data set was expanded based on own measurements.

The comparison of the clinker from SM(P)/2% CaCl₂ with the simplified calculated average pd-AAC composition SC-pd-AAC / 2 wt% CaCl₂ shows a general agreement. Deviating phase contents are due to an incomplete reaction. The experimental sample contains two C₂S modifications (61.4 wt% β-C₂S, 5.4 wt% α-C₂S, total 66.8 wt%). Assuming that the amorphous phase (15.2 %) comprises a proportion of amorphous C₂S that corresponds in mass to the content of C₂S in the crystalline phase (66.8 wt%), the total content of C₂S in the sample is 76.9 wt%. This C₂S content is slightly reduced compared to the calculated composition (85.2 wt%) due to the incomplete turnover of the raw materials quartz, calcite, lime, and anhydrite. The basic agreement of experiment and calculation enables the use of thermodynamic data to calculate mass and energy balances.

2.6. Comparing RC-BCC and Portland cement clinker

The RC-BCC technology is in the stage of basic validation in a laboratory or relevant environment (TRL 4–5). A comparison with PCC is only possible to a limited extent. This section examines whether, and if so, which types of RC-BCC could be suitable as substitutes for PCC, whether they meet the requirements standardized for PCC and cement, and whether adaptation of standards might be necessary.

Table 2

Measured content of main clinker minerals in RC-BCC prepared from the indicated pd-AAC, limestone, and mineralizer in wt%. For comparison, the calculated phase content of the standard clinker SC-pd-AAC is given. Estimated standard deviations (E.s.d.s) of Rietveld refinement are provided in parentheses. XRA: x-ray amorphous; T: Terneite; Q: Quartz; C: Calcite; L: Lime (CaO); A: Anhydrite (Ca₂SO₄); W: Wollastonite (CaSiO₃); E: Ellestadite; NS: Na₂SO₄. Difference from 100 % in measured samples: minor phases (not shown).

pd-AAC / mineralizer	XRA	α'H-C ₂ S	β-C ₂ S	T	Q	C	L	A	W	E	NS	Sum
SM(P) / -	-	0	90.0 (6)	0.44 (13)	2.52 (7) 7)	0.63 (11)	-	-	0.14 (5)	-	-	93.73 (99)
SM(D1) / -	10.2 (1.7)	4.2 (3)	47.2 (9)	1.17 (11)	8.76 (16)	7.02 (15)	12.07 (17)	-	4.32 (12)	-	-	94.94 (2.52)
SM(D2) / -	4.8 (1.3)	1.08 (1.7)	62.6 (6)	11.36 (17)	5.62 (12)	1.49 (11)	2.52 (5)	4.50 (9)	0.64 (10)	-	-	94.61 (3.70)
SM(D3) / -	-	1.35 (18)	65.4 (6)	8.31 (16)	5.57 (13)	1.23 (12)	1.84 (5)	7.46 (11)	0.99 (11)	-	-	92.15 (92)
SM(P) / 2 % CaCl ₂	15.0 (7)	-	63.8 (4)	0.12 (7)	2.11 (4)	-	0.17 (5)	0.43 (6)	0.6 (1)	9.9 (1)	-	92.13 (1.16)
SC-pd-AAC / 2 % CaCl ₂	-	-	85.2	-	-	-	1	-	-	13.8	-	100.0
SM(P) / 5 % Na ₂ CO ₃	22.1 (9)	7.6 (2)	61.3 (4)	-	0.22 (4)	0.07 (4)	-	-	1.0 (2)	-	1.8 (4)	94.6 (2.18)

According to the definition given for PCC in DIN EN 197-1:2011, the processing of RC-BCC and PCC does not differ significantly. In particular, the standard does not define a minimum process temperature. As an essential requirement, PCC shall consist of at least two-thirds by mass of calcium silicates (C₃S and C₂S), the remainder consisting of aluminum and iron-containing clinker phases and other compounds. The ratio by mass (CaO)/(SiO₂) shall be not less than 2.0. The content of magnesium oxide (MgO) shall not exceed 5.0 wt%.

Additional chemical requirements apply to certain types of cement that contain PCC, e.g. masonry cement (DIN EN 413-1:2011), very low heat special cement (DIN EN 14216:2015), Portland composite cement CEM II/C-M, Composite cement CEM VI (DIN EN 197-5:2021), super-sulfated cement (DIN EN 15743:2015). They mainly concern the loss on ignition (LOI) and insoluble residues (typically < 5 wt%) as well as the sulfate and chloride contents. Contents are analyzed according to DIN EN-196-2:2013. While RC-BCC meets the required properties in terms of LOI and insoluble residue, its sulfate and chloride contents vary on the secondary raw materials used, the mineralizer, and the chemical stability of the mineral phases formed. The maximum tolerable sulfate content, typically 3.5 to 4.5 wt% SO₃, can also be maintained by blending with PCC.

On the other hand, the use of (earth) alkali chloride as mineralizer is critical for standardized applications due to chloride-induced corrosion of reinforcements in structural components. Typically, the chloride content is limited to 0.1 wt%. High chloride contents are only tolerated in special mortars, according to DIN EN 413-1:2011.

The typical application for high-belite clinker in China, very low-heat cement, is not standardized within the European framework. Instead, according to DIN EN 14216:2015, high amounts of ground granulated blast-furnace slags are combined with PCC for this purpose.

The performance of RC-BCC in AAC processing has been successfully tested on the lab scale (up to 50% substitution of PCC) and on an AAC production line (industrial scale, 25 % substitution of PCC, Stemmermann et al. 2022, 2023). The setup is described in Section 3.2. Technically, RC-BCC was ground to 90 wt% passing through a sieve of 90 μm. Subsequently, fixed ratios of RC-BCC and OPC were mixed with the other raw materials quicklime, anhydrite, primary sand, aluminum paste or powder, and water to form a thin mortar that was further processed into AAC by the industrial partner Xella (Stemmermann et al. 2022). The used recipes correspond to commercial products. The joint mass of RC-BCC and OPC replaced the cement content of the recipe.

2.7. Estimation of material balances, energy consumption, and CO₂ emissions

Based on the simplified formulation for the standard clinker SC-pd-

AAC given in Tables A-1 and A-2 (supporting information), mass balances for the production of RC-BCC, including fuel and combustion gasses for different firing conditions, were estimated. Table 3 lists calculated energy balances for all investigated variants. In the case of oxyfuel combustion, additional energy is accounted for oxygen generation. A value of 0.5 kWh per standard cubic meter (SCM) has been taken from a Vacuum-Pressure Swing Adsorption (VPSA) installation with 100 SCM/h and 80% oxygen purity (Banaszekiewicz and Choroski, 2018). The maximum product temperature is assumed to be 1000 °C.

The limestone-pd-AAC mixing ratio, the relative amounts of CO₂ emission, and the clinker quantities rely on the composition of the pd-AAC fines. The following Sankey diagram (Fig. 2) gives a graphical representation of the mass flows and dependencies.

Pd-AAC consists of tobermorite, quartz, gypsum and water; all amounts of silicon dioxide and calcium oxide are considered reacting. Pure calcium carbonate is added to adjust belite chemistry: for pd-AAC a typical molar CaO-SiO₂ ratio is 0.7. Pd-AAC samples usually contain up to 5 wt.% of additional oxides of Al, Fe, alkalis and earth alkalis, which are inert, thus not considered here. Hydrated calcium chloride is added as mineralizer.

The resulting RC-BCC leaving the rotary kiln contains belite and ellestadite, the later containing all the sulfate and chloride. The gas phase is composed of carbon dioxide and water vapor.

Thermal efficiencies were estimated based on PCC processing. Thermodynamically, the production of PCC requires approx. 1800 kJ/kg clinker (Locher 2002). Due to thermal losses by off-gas, wall losses, cooler exhaust air, and clinker, which decrease in this order, the actual thermal energy demand, according to BAT (European Commission 2013) for PCC increases to 3060–3620 kJ/kg clinker if no waste is used as fuel. These values include start-ups and shutdowns and result in a thermal efficiency of 50–60%, which may decrease when waste is used. Recent data from the German cement industry (VDZ ed., 2022) result in an energy input of 3875 kJ/kg (thermal efficiency 46%), taking into account the current clinker factor (0.73).

Plants for the production of a belite clinker do not yet exist. A hypothetical comparison of the thermal efficiency of the technology with the production of PCC shows great advantages for the losses via off-gas, especially for electric heating, since the quantity and temperature of the off-gas are significantly reduced. This reduction also applies to a limited extent to the oxyfuel technology. The lower firing temperature additionally reduces wall losses, losses from cooler exhaust air, and clinker. However, the lower maturity of the belite technologies has a negative impact. While air-operated gas burners are mature, the oxyfuel technology is in the optimization phase. Large-scale electric kilns at an operating temperature of 1000 °C are a technological challenge. For air-

Table 3

Calculated mass and energy balances of the production of 1t RC-BCC from the raw materials SC-pd-AAC / 2 wt% CaCl₂ (Table A-2) for an all-electric process, for methane combustion equipped with oxyfuel and for air-operated methane combustion including energy commodities and oxygen/air. The CO₂ emissions from PCC are given for comparison, not differentiating biogenic or fossil fuels. (process emissions + fuels, = direct CO₂ emissions cement / clinker factor (VDZ ed., 2020, VDZ ed., 2022)).

Mass balance							
	Educts RC-BCC [kg/t]	Products RC-BCC	[kg/t]	H ₂ O, gases, cumulated	CO ₂ (process + fuels) [%]		
Electric heating	AAC	607	RC-BCC	1000			
	Tobermorite	363	Belite	852			
	Quartz	144	Ellestadite	138			
	Water	31	CaO	10			
	Gypsum	69					
	CaCO ₃	840	H ₂ O	107	107		
	CaCl ₂ .6H ₂ O	34	CO ₂	369	369	45%	
	Sum	1482	c(CO ₂) off-gas: ca. 100%	1476			
+ Oxyfuel	+CH ₄	47	+H ₂ O	103	210		
	+O ₂	197	+O ₂	9	9		
			+CO ₂	132	502	61%	
	Sum	1725	c(CO ₂) off-gas: ca. 95%	1721			
+ Gas combustion in air	+CH ₄	12	+H ₂ O	24	235		
	+O ₂	49	+O ₂		9		
	+N ₂	800	+N ₂	800	800		
			+CO ₂	38	540	66%	
	Sum	2586	c(CO ₂) off gas: ca. 36%	2584			
PCC (for comparison)		CO ₂	820			100%	
Energy balance							
	Thermal efficiency*	Heat demand [kJ/kg]	El. heat [kWh/t]	Milling [kWh/t]	Oxygen generation [kWh/t]**	Oxygen [SCM/t]	Sum El. supply [kWh/t]
Electric heating	60%	1937	538	110	-	-	648
+ Oxyfuel	50%	2606	CH ₄	110	69	700	179
+ Gas combustion in air	60%	3243	CH ₄	110	-	•	110
	*: Estimate)				**: Assumption 0.5 kWh /SCM)		

operated gas burners as a mature technology, a thermal efficiency of 60%, equal to the best BAT cement plants, was assumed (also reflecting the reduced temperatures compared to OPC clinkering). Due to the lower technical maturity of the oxyfuel technology, the estimated efficiency was reduced to 50%. For electric heating, 60% efficiency was assumed, as strongly reduced heat losses presumably compensate for the lower technical maturity. Concerning energy efficiency, the scaling of the plants is also essential but was not considered due to a lack of data. The electricity required for grinding the raw materials and products as well as for process control, fans, and transport, was not determined separately. Instead, a standard value of 110 kWh/t clinker was adopted from the production of OPC (VDZ ed., 2020).

3. Life cycle assessment of closed-loop recycling of pd-AAC to RC-BCC

3.1. Goal and scope

The LCA of RC-BCC production from pd-AAC and the AAC

production using RC-BCC is done with the LCA software openLCA (GreenDelta 2019) and the database ecoinvent 3.8 cut-off (ecoinvent 2021). The goal is to determine whether closed-loop recycling of AAC via RC-BCC production is environmentally beneficial compared to landfilling.

Instead of the classical cradle-to-grave approach assessing primary resource extraction, production, use phase, and end-of-life, the following LCA of RC-BCC production from pd-AAC adjusts the system boundaries and the functional unit using the so-called zero burden approach (Nakatani 2014). It excludes efforts for primary resource extraction, production, and use as well as transports. So, the pd-AAC enters the assessment of the end-of-life stage without any burdens. This assumption is reasonable since we only want to assess and compare waste treatment options at the end of life. The system boundaries include pd-AAC at the demolition site, RC-BCC production, AAC production using RC-BCC, and transports in between (see Fig. 1). This simplification does not influence the comparison of different end-of-life options, as the excluded efforts would be the same for all. Instead of the production output, the system's input (1 kg pd-AAC) is the functional

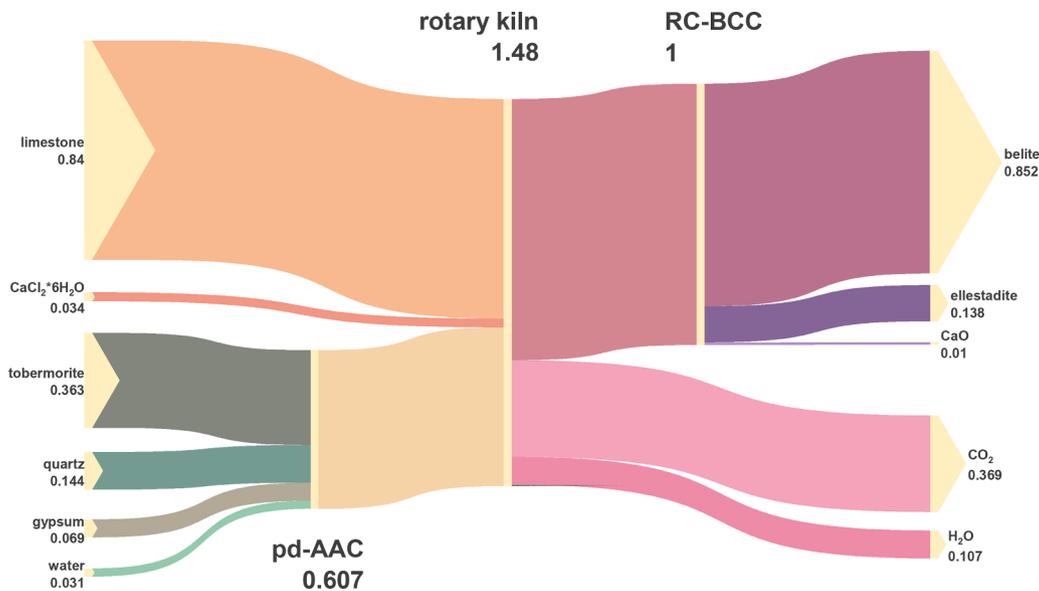


Fig. 2. Sankey diagram of mass-flow for belite clinker production based on pd-AAC input with C/S ratio = 2, normalized for RC-BCC.

unit. It allows for comparing different end-of-life options with different final products. A system expansion covers the handling of sorting residues following DIN EN ISO 14040:2021-02 and DIN EN ISO 14044:2021-02.

Furthermore, the avoided burden approach is applied to consider the final products' value (Nakatani, 2014). It allows rewards for avoided primary production of the replaced product. DIN EN ISO 14040:2021-02 and DIN EN ISO 14044:2021-02 name the system expansion as the best way to deal with valuable outputs and wastes in an LCA. It's assumed that the AAC produced using RC-BCC replaces primary AAC. Thus, the LCA includes an environmental reward since burdens from primary AAC production are avoided. This LCA reflects the specific case of Central Europe/Germany, so the German electricity mix is chosen for impact assessment.

3.2. Life cycle inventory (LCI) analysis

Here, all processes, flows, data sources, and assumptions for the LCA are described. All accompanying inventory data, uncertainties and references are listed in the Supporting Information S2. Data from ecoinvent 3.8 is used for assessing the pre-treatment processes of crushing, grading, and purifying, as well as for landfilling and primary production. The ecoinvent data quality system is used for Monte Carlo simulation to conduct a sensitivity analysis.

Today, landfilling is the main end-of-life option for pd-AAC and is the benchmark. The pd-AAC landfilling is calculated using the ecoinvent 3.8 dataset "treatment of inert waste, inert material landfill | inert waste, for final disposal", considering energy efforts for landfilling and occupation and transformation efforts. Furthermore, purifying residues from the pd-AAC recycling process are assumed to be landfilled and accounted for similarly.

The crushing is assessed with the ecoinvent 3.8 dataset "rock crushing | rock crushing". Material losses in the crushing process are marginal and not considered. Transports from the demolition site to a recycling plant are assumed to be 50 km. Pd-AAC can contain different adhesions or impurities, which should be minimized in the recycling process through purifying. Furthermore, a grading process can separate fine pd-AAC powder from coarse pd-AAC granulate. Only the powder is used for RC-BCC production. The coarse granulate could be used for other recycling options (Section 1) or recirculated and crushed again to receive pd-AAC powder. The ecoinvent 3.8 dataset "treatment of waste brick, sorting plant | waste brick" is used to assess a combination of both

processes. Datasets for crushed AAC do not exist, and primary data from industrial sites are unavailable. Furthermore, 0.01 kg purifying residue per kg purified pd-AAC is assumed to be sorted out while the grading has no material losses. Efforts are allocated by mass to the two final products: pd-AAC powder and granulate.

RC-BCC production is assessed using primary data for energy and mass flows (Section 2) in four energy supply variants: natural gas, oxyfuel, conventional electricity (German electricity mix), and 100 % renewable electricity. AAC production using RC-BCC is calculated for different AAC density classes (AAC-0.35, AAC-0.5, and AAC-0.55). The main inputs for AAC production are sand, cement, quicklime, and anhydrite. Production recipes are taken from Volk et al. (2023). Cement is substituted by 25% or 50% RC-BCC (Table A-3). The German electricity mix is available as an ecoinvent dataset. 100 % renewable electricity is represented by the dataset "market for electricity, medium voltage, renewable energy products | electricity, medium voltage, renewable energy products" which is only available for Switzerland. The transport distance between RC-BCC production and the AAC production plant is assumed to be 50 km.

3.3. Life cycle impact assessment

The life cycle impact assessment (LCIA) is performed with the "ReCiPe 2016 Midpoint (H)" method. The cultural perspective of Hierarchist (H) is preferred to Individualist (I) and Egalitarian (E) because it reflects common assumptions, e.g., the climate change based on 100 years horizon instead of 20 years (I) and 500 years (E). All included midpoints are presented in Table A-4, and their assessment results are in Figures A-1, A-2, and in supporting information B.

In the RC-BCC production, pd-AAC processing (green) and transport efforts (blue) are relatively low and hardly visible for any midpoint (Fig. 3). In contrast, energy efforts for electricity (yellow) or natural gas (gray) influence the results significantly. Process emissions due to natural gas combustion and direct CO₂ emissions from limestone contribute extensively to global warming. Direct process emissions do not affect other midpoint categories, as only direct CO₂ emissions are considered (Fig. 1). As expected, renewable electricity usage in the rotary kiln has the lowest impact for all considered midpoints. The global warming (GW) of RC-BCC can be reduced from 0.76 kg CO₂-Eq/kg RC-BCC (German electricity mix) / 0.66 (oxyfuel) / 0.67 (natural gas) to 0.40 kg CO₂-Eq/kg RC-BCC (100% renewable electricity). However, natural gas firing (atmospheric or oxyfuel combustion) leads to lower impacts

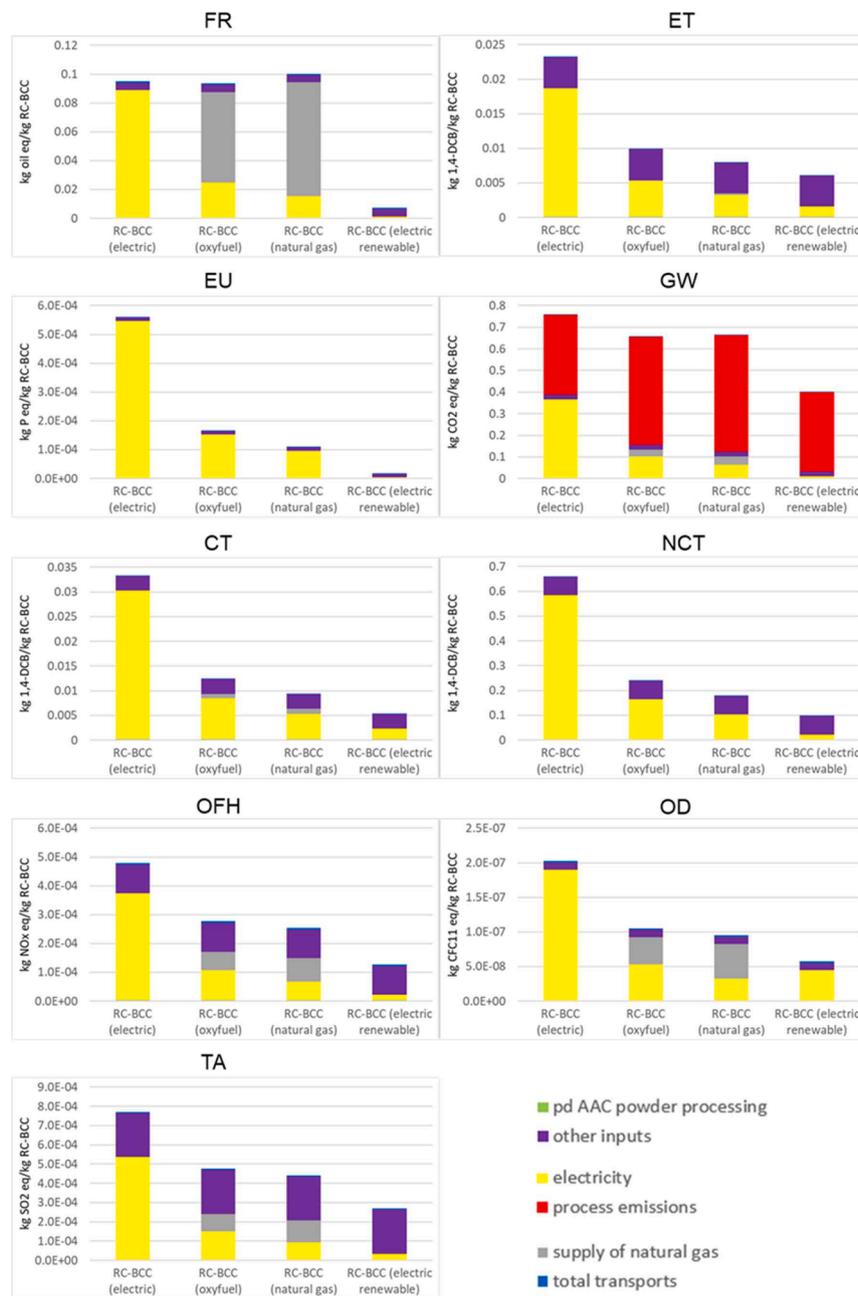


Fig. 3. LCA results for RC-BCC production with different firing conditions (FR: Fossil resource scarcity, ET: Freshwater ecotoxicity, EU: Freshwater eutrophication, GW: Global warming, CT: Human carcinogenic toxicity, NCT: Human non-carcinogenic toxicity, OFH: Ozone formation–Human health, OD: Stratospheric ozone depletion, TA: Terrestrial acidification).

than using the German electricity mix for almost every midpoint, as the German electricity mix still depends significantly on fossil resources.

Fig. 4 shows the results for AAC production using RC-BCC to substitute either 25 % or 50 % of the OPC. Overall, the substitution can reduce the impacts of AAC production. Concerning the GW, savings are highest for substituting OPC with RC-BCC produced using 100 % renewable electricity, followed by oxyfuel combustion, natural gas, and conventional electricity. This order corresponds to the LCA results of the RC-BCC production and is the same for all AAC density classes. Global warming, e.g., is reduced from 0.01 kg CO₂-Eq/kg pd-AAC (landfilling, status quo) to -0.76 kg CO₂-Eq/kg pd-AAC (substitution of OPC by RC-BCC with 100 % renewable electricity).

The substitution amount (pd-AAC input) hardly influences the total savings as the results are normalized to the functional unit of 1 kg pd-AAC. However, total savings are higher in the 50 % than in the 25 %

substitution scenario if calculated per kg AAC-0.35 (final product). The GW of AAC-0.35 is reduced from 0.52 kg CO₂-Eq/kg (primary production) to 0.45 kg CO₂-Eq/kg (50 % substitution, RC-BCC with 100 % renewable electricity). Results for AAC-0.5 (reduction from 0.45 to 0.40 kg CO₂-Eq/kg) and AAC-0.55 (reduction from 0.39 to 0.35 kg CO₂-Eq/kg) are comparable to AAC-0.35. Both overall impacts and savings are somewhat lower for AAC-0.5 and AAC-0.55 as their production needs less cement than AAC-0.35.

3.4. Interpretation

The LCA results show significant potential for savings in several environmental impact categories, especially considering GW. Savings could reach 0.77 kg CO₂-Eq/kg pd-AAC compared to the status quo (landfilling) when RC-BCC is produced using 100% renewable

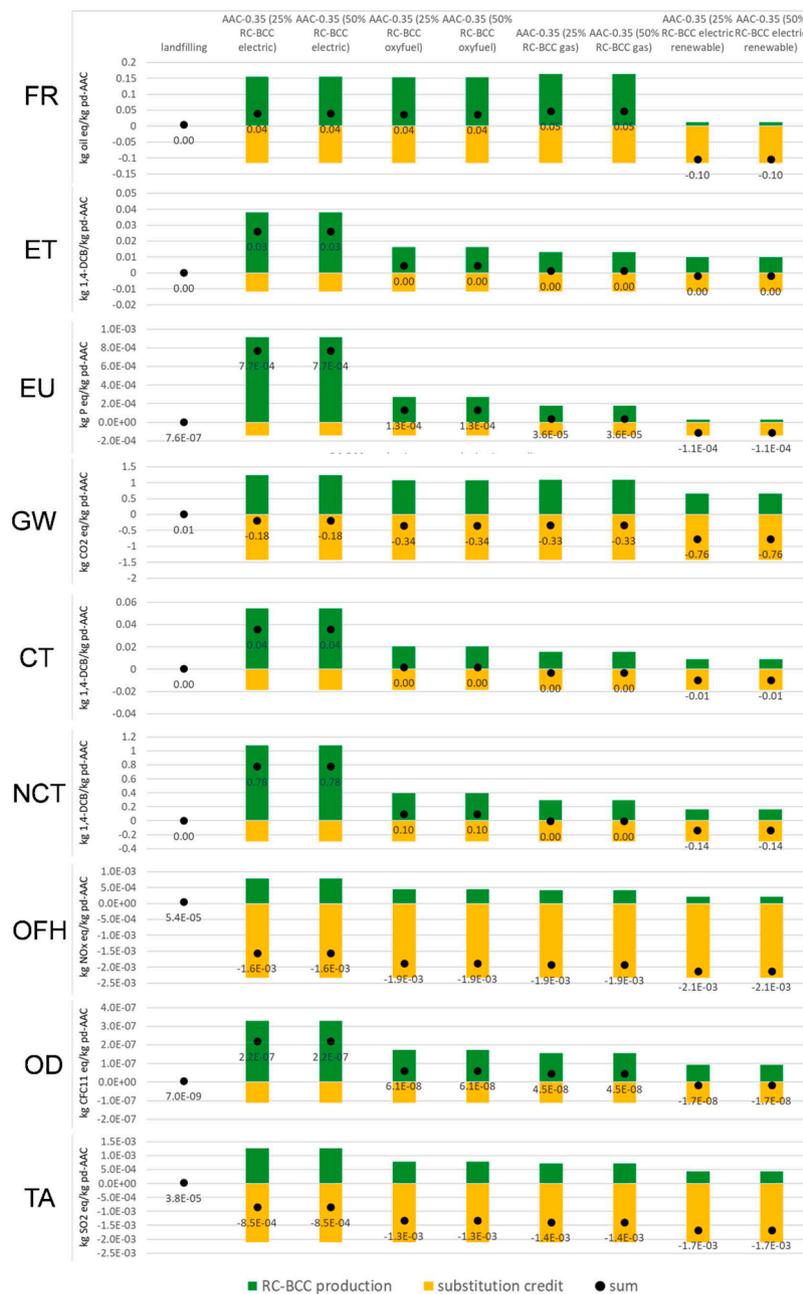


Fig. 4. LCA results for the AAC-0.35 production with RC-BCC content. Savings in relation to the functional unit 1 kg pd-AAC are displayed for different midpoints (FR: Fossil resource scarcity, ET: Freshwater ecotoxicity, EU: Freshwater eutrophication, GW: Global warming, CT: Human carcinogenic toxicity, NCT: Human non-carcinogenic toxicity, OFH: Ozone formation–Human health, OD: Stratospheric ozone depletion, TA: Terrestrial acidification).

electricity and OPC is substituted in AAC production. Firing the process with oxyfuel or natural gas still reaches savings of about 0.34 to 0.35 kg CO₂-Eq/kg pd-AAC. Impacts of 1 kg AAC-0.35 can be reduced by 0.07 kg CO₂-Eq/kg. This reduction of 13.5 % is significant, considering only 15.5 % of the overall input material is substituted. However, many midpoints show higher impacts when the RC-BCC is produced using conventional electricity.

A comparison to other recycling options for pd-AAC investigated by Volk et al. (2023) shows that RC-BCC produced with 100% renewable electricity shows the highest CO₂-Eq saving potential. However, RC-BCC produced with natural gas or oxyfuel combustion would be outperformed by direct usage of pd-AAC powder in AAC production and some open-loop recycling options like light mortar production.

German AAC production is around 3.5 million m³ (GENESIS 2022), of which AAC-0.35 accounts for about 45 %, AAC-0.5 for 20 %, and

AAC-0.55 for 10 % (Volk et al. 2023). More than 82,000 t of pd-AAC could have been recycled in a closed loop, assuming a 50 % substitution of OPC with RC-BCC in AAC-0.35, AAC-0.5, and AAC-0.55. This amount equals 12 % of Germany’s expected total pd-AAC amount of 700,000 t in 2022 (Steins et al. 2022). Total savings in greenhouse gas emissions would sum up to 63,600 t CO₂-Eq annually if the RC-BCC is produced using renewable electricity and 28,200 t CO₂-Eq if the RC-BCC is produced using natural gas.

3.5. Sensitivity analysis through Monte Carlo simulation

A Monte Carlo simulation with 10,000 runs for each process is performed for all scenarios (Table 1-3) to analyze the sensitivity of the LCA results. The ecoinvent data quality system determines the parameters of a lognormal uncertainty function. It includes data reliability,

completeness, temporal correlation, geographical correlation, and further technological correlation. Uncertainty values for these categories are taken from the ecoinvent 3.8 database if possible. Inputs for the RC-BCC production are rated 1 (data reliability), 5 (completeness), 1 (temporal correlation), 1 (geographical correlation), and 5 (further technological correlation) to reflect the presented situation. The simulation shows that the initially calculated values are robust (Fig. 5 and supporting information B). Median values for all scenarios and all midpoints are close to the original value. The RC-BCC production shows sensitivities of around -10 % to -20 % and +20 to +30 %, depending on the firing variant. The final AAC production with RC-BCC shows even lower sensitivities of less than -10 % and up to +10 %. The highest sensitivity for all scenarios and midpoints is around -20 % to +40 % in the 25 %/75 % percentiles (supporting information B). Overall, calculation results seem robust even though the RC-BCC production at the current TRL is associated with some data uncertainty, especially for completeness and technological correlation.

4. Discussion

Processing RC-BCC in general and in particular from pd-AAC is new; details are presented here and in Stemmermann et al. (2022). Significant amounts of RC-BCC have been produced by a service provider and used in a semi-industrial test in AAC production, where RC-BCC successfully replaced 25 % of OPC. The LCA is based on our experimental data. However, the process has a TRL of 4–5, only optimized on the laboratory scale.

RC-BCC shows clear advantages concerning energy consumption and CO₂ emissions compared to PCC. However, the technology also has disadvantages and risks; e.g. at least to date, hydraulic reactivity is significantly slower, which may result in longer processing times for products or the need for accelerators, thus higher costs.

Other critical issues are homogeneity and quality of the product if variably composed and contaminated secondary raw materials are processed. High levels of impurities result in a reduced proportion of hydraulically effective clinker minerals. Another difficulty is the permanent supply of distributed residual materials of defined quality for clinker production and its transport logistics compared to quarrying raw materials. Work on technical applications of the RC-BCC and the generated CO₂ is in progress but remains largely unresolved. Legal framework conditions are lacking.

For future assessments, two cases are relevant: (1) Exclusive processing of pd-AAC and (2) Blending of pd-AAC with other materials

(fiber cement (cellulose-based), waste concrete). The latter might increase plant utilization and help to increase recycling material flows but would require adjustments of raw meals and process parameters.

The assessment results are associated with considerable uncertainty. The process shall be further developed to a higher TRL with more precise process parameters to reduce these uncertainties. However, the LCA data's uncertainty is somewhat included via Monte Carlo simulation (Section 3.5).

Moreover, the scaling and placement of plants have to be discussed. Large plants are cost-efficient due to economies of scale (Norman 1979) and energy-efficient due to reduced heat losses. Comparing the energy demand in BAT (3000 t/d, 3,340 kJ/kg, mean) and average German plants (2675 t/d, 3,875 kJ/kg; energy average of all kilns, capacity for rotary kilns only, 98.8 % of the total capacity, VDZ ed., 2022) suggests that other factors play a significant role. One factor is the increasing energy demand when using secondary wastes as fuel. If waste fuels are combined with carbon capture usage and storage, indirect heating options would be required not to contaminate the concentrated CO₂. Moreover, transport and reverse supply chain problems might arise in a large central plant. Given the high processing temperature (1450 °C), small-scale PCC processing seems complicated.

RC-BCC production on a small scale (125 t/d) seems technologically feasible. However, data is lacking, especially for various heat input options. Appropriate approaches are currently being developed. Small-scale RC-BCC production would require many more plants with smaller sourcing areas, stocks and transport distances. Moreover, they could face material shortages if sourcing regions become too small. Large and small plants must adapt to the volatile and seasonal demolition, e.g. via stockholding.

Besides, the assumption of 110 kWh/t for milling of pd-AAC might be too conservative since the material is quite soft. Moreover, the LCA results might change due to efficiency gains, significantly higher pd-AAC volumes in the future, higher belite amounts in recipes, or changing recycling strategies. It is striking how much the choice of electricity mix (German electricity mix or 100 % renewables) influences the results. In the case of oxyfuel/natural gas, the German electricity mix was used to assess electricity efforts. When changing this to 100 % renewable electricity, the impact of the oxyfuel process would be reduced more than of the normal combustion since additional electricity is needed for oxygen production. Economic aspects, market barriers, or further process integration are not assessed.

AAC production using RC-BCC is considered in different scenarios, combining two substitution levels and three AAC density classes. The

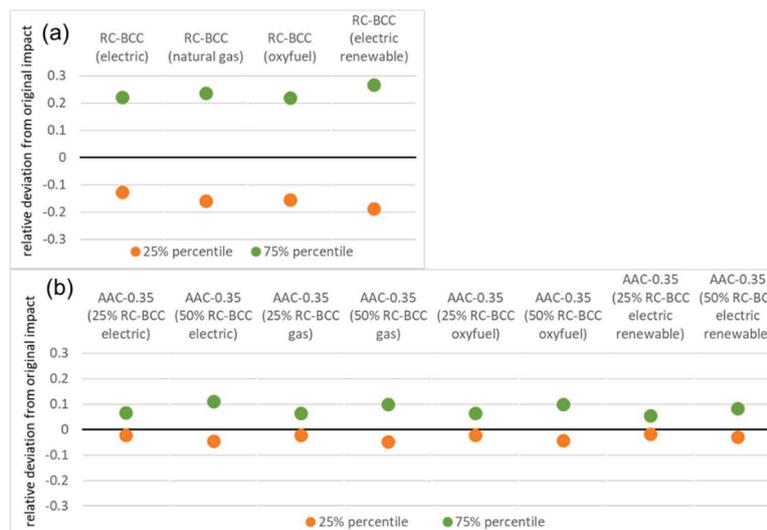


Fig. 5. Sensitivity of the LCA results in a Monte Carlo simulation (25 % and 75 % percentile) for all considered scenarios, including RC-BCC production (a) and final AAC production (b), for the midpoint GW.

density class influences the composition of the input materials and, therefore, affects the results. However, the substitution levels do not influence the results since the functional unit is 1 kg pd-AAC. Total savings per kg produced AAC are higher in the 50 % substitution scenario. Since pd-AAC volumes are expected to rise significantly in Germany and Europe (Steins et al., 2021, 2022), AAC recycling should aim at high substitution rates to minimize environmental impacts.

5. Conclusion

Using a small semi-industrial kiln RC-BCC from waste concrete has been successfully processed. The suitability of RC-BCC as a partial cement substitute in aerated concrete production has been demonstrated in technical trials (Stemmermann et al. 2022). The processing of RC-BCC from pd-AAC was established on a laboratory scale using samples from real collected waste with different levels of contamination. Both the specific energy requirement and the specific emissions of CO₂ can be minimised if electrical heating based on renewable electricity is used in the production process. For economic reasons, however, plants with an annual production of at least 50 kt are necessary. An electrically heated rotary kiln intended for upscaling the technology on this scale is uncharted technological territory. Further difficulties of upscaling concern the control of a homogeneous raw meal composition, heat recovery for high energy efficiency and the constant supply of suitable primary and secondary raw materials.

Improvements in various environmental aspects through pd-AAC recycling are possible, especially concerning CO₂-Eq savings and landfill capacity. Closed-loop pd-AAC recycling via the belite route and substituting OPC shows promising results and significant potential savings of 0.77 kg CO₂-Eq/kg pd-AAC compared to the status quo (landfilling) by using renewable electricity. Savings reach 0.35 kg CO₂-Eq/kg pd-AAC when using oxyfuel combustion or natural gas firing. The gained reduction of 13.5 % is significant, considering that only 15.5 % of the overall input material is substituted. The results indicate that closed-loop recycling of pd-AAC could significantly reduce environmental burdens associated with the current landfilling. However, the assessment also shows some worsening of environmental impact categories, e. g., freshwater ecotoxicity or eutrophication, human carcinogenic toxicity, and stratospheric ozone depletion when using the German electricity mix. Other recycling options are more promising regarding these environmental impact categories (e.g. Volk et al. 2023).

Future research should focus on assessing a pilot plant instead of laboratory data to enhance decision-making. Furthermore, a system analysis and network design for a full AAC circularity could help to identify the optimal recycling routes, plant capacities, and placements under given or future conditions, e.g. spatial and temporal availability of rising pd-AAC, quality aspects and current regulation.

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Supporting Information

Supporting Information A: This supporting information (A) provides additional figures and tables for the main article including the chemical composition of pd-AAC and mass balance mixtures and an overview on considered scenarios in the LCA. Moreover, the assessed midpoints are listed and the graphic results of the impact assessment of landfilling and processing pd-AAC for recycling as well as various pd-AAC recycling options are displayed.

Supporting Information B: This supporting information (B)

provides the full LCA information in tabular form (MS Excel format). It is the basis for Figures 3 and 4 in the manuscript as well as for Figures A-1 and A-2 in the supporting information A.

CRedit authorship contribution statement

P. Stemmermann: Writing – review & editing, Writing – original draft, Investigation, Funding acquisition, Data curation, Conceptualization. **R. Volk:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **J.J. Steins:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation. **G. Beuchle:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Data curation.

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

The data that supports the findings of this study are available in the supporting information of this article.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.resconrec.2023.107404](https://doi.org/10.1016/j.resconrec.2023.107404).

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