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Drava river sediment in clay brick production: Characterization, properties, and environmental performance

Mojca Božič [a], Lea Žibret [b], Davor Kvočka [b], Alenka Mauko Pranjić [b], Boštjan Gregorc [a], Vilma Ducman [b,*]

[a] *Dravske elektrarne Maribor d. o. o., Obrežna ulica 170, 2000, Maribor, Slovenia*
[b] *Slovenian National Building and Civil Engineering Institute, Dimičeva Ulica 12, 1000, Ljubljana, Slovenia*

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ABSTRACT

The ever-growing worldwide demand for fired clay brick has resulted in the shortage of clay in many parts of the world. Therefore, there is a need to look for more sustainable alternative materials for the brick manufacturing. This study has investigated the potential use of the untreated Drava River sediment as a substitute material for clay in the production of fired bricks, with the research being conducted at both laboratory and industrial level. At the laboratory level, brick specimens were prepared by mixing clay with different river sediment proportions (ranging from 10 to 50 wt%) and were fired at 950 °C, with microstructural and various physical–mechanical properties being analyzed. Elevated carbonate content in Drava river sediment results in higher weight loss during firing at temperatures up to 950 °C, comparing to firing pure brick-making clay. Consequently, the addition of sediment increases porosity of fired bricks, which results in lowering of their mechanical properties. Results reveal that the compressive strength of the pure clay sample was 79.5 MPa, while the compressive strength of the sample with the addition of river sediment from 10 wt% to 50 wt% decreased from 73.9 MPa to 26.2 MPa, respectively. Despite the lower compressive strength, the 26.2 MPa is still above the limit value of 10 MPa specified in the standard EN 772–1 [1]. At the industrial level, hollow clay bricks were prepared with 20 wt% of the river sediment and fired in a tunnel kiln. Inclusion of the river sediment also decreased compressive strength from 38 MPa for pure mixture to 26 MPa for 20 wt% of the sediment addition, confirming usability of Drava sediment in brick production. In addition, LCA study has been conducted to evaluate the environmental impacts associated with the industrial production of classic bricks and bricks with the addition of the river sediment. The obtained results have shown that the bricks made with the addition of the Drava River sediment are sustainable and environmentally friendly and meet all the requirements specified in the relevant regulatory standard.

1. Introduction

The awareness that natural resources are limited has influenced our decision to gradually shift from linear model of economy (“take-make-dispose”) to the circular model of economy (“reduce-reuse-recycle”) [2]. The circular economy concept is considered as one of the key frameworks for reducing the environmental impact of the construction sector [3]. For example, the construction sector consumes more than 50% of all natural resources available in Europe and is responsible for more than 30% of the European carbon

* Corresponding author.
E-mail address: vilma.ducman@zag.si (V. Ducman).

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footprint [4]. Therefore, the construction sector has to become more sustainable in order to reduce the enormous consumption of natural resource and corresponding waste generation. This can be achieved by using the so-called waste materials [5–7].

A type of an inert waste that can be used in various branches of construction sector is sediment [8]. Sediment is a solid material that is moved and deposited within the rivers, dams, reservoirs, canals and harbours [9]. Millions of tons of sediment are dredged every year, with the environmental dredging being generally used for the reduction of sediment loads in rivers [10]. However, as the sediments have the tendency to absorb contaminants (e.g. heavy metals), they present a serious challenge for river managers [11]. Therefore, numerous researches have investigated the potential of the use of sediments for different construction applications in order to find sustainable and environmentally friendly alternative to the disposal of the dredge sediment on landfills and open waters [12]. Based on the chemical and mineral composition, the sediment can be used in: (i) brick production [13–15], (ii) clinker production [16], (iii) mortars/concretes as fillers or aggregates [17], (iv) light aggregates manufacture [18], (v) supplementary cementitious materials [19], and (vi) earthworks, such as soil stabilization, revitalization, backfills etc. [20].

Due to the ever-growing worldwide brick production, there is already a shortage of clay in many parts of the world [21]. For example, to protect the raw clay resources and the environment, some countries (e.g. China and India) have already started to limit the use of bricks made from clay [2,22]. Hence, the brick-making sector is constantly looking for suitable additives or substitutes for raw clay [23–27]. The research in the field of the use of secondary raw materials to partially or fully replace the clay in the brick-making process has been on the rise in recent years thanks to the adoption of the circular economy principals within the industry [28,29]. Consequently, numerous alternative building materials have been developed in order to boost the sustainable development and reduce the environmental impacts [30]. Moreover, the latest brick discoveries also address the new methods of brick processing for increasing the energy efficiency [31,32].

Life Cycle Assessment (LCA) is a widely used methodology to evaluate the potential environmental impacts associated with the production, use and end-of-life processes of construction materials [33,34]. A typical LCA study consists of four phases (goal and scope definition, inventory analysis, impact assessment, interpretation of results) and is conducted in accordance with the principles and framework for LCA defined in the international standard for LCA ISO 14040 [35] and ISO 14044 [36]. LCA method has been used to evaluate different life-cycle stages of the fired clay bricks and different substitute materials [37–39]. In general, the results indicate that the production stage contributes the most to the overall environmental footprint and that environmental benefits can be achieved by using both organic and inorganic waste materials [40].

The use of river sediment as a material in the brick manufacturing has been investigated by several authors [41–48]; however, these studies have evaluated chemical and mechanical properties of bricks on a laboratory scale, whereas they have not considered industrial production and thus the related environmental parameters (e.g. exhaust gases concentrations). As only a handful of studies has reported on successfully conducted industrial experiments [49–51], it is not surprising that the use of the dredged river sediments as a secondary material in the brick manufacturing process is not widely applied on a commercial scale [52].

This study has investigated the potential use of the untreated Drava River sediment as a substitute material for clay in the production of fired bricks, with the evaluation process being conducted at both laboratory and industrial scale. Two different sediment samples have been considered in order to determine the impact of the sediment ageing on brick's characteristics, i.e. (i) sediment that has been naturally dehydrated for one year and (ii) sediment that has been naturally dehydrated for more than 10 years. In the first stage of the study, the chemical and mineralogical analysis of the raw materials has been conducted, with materials later being mixed in different sediment/clay proportion and fired at different temperatures. The ceramic-technological tests have been performed on brick samples to determine the linear shrinkage, water absorption, bending and compressive strengths, bricks' density and porosity. In the second stage of the study, an industrial test has been conducted based on the optimal sediment/clay proportion determined in the laboratory, with different production parameters being monitored and analyzed. Finally, the environmental impacts associated with the industrial production of both traditional and alternative brick mixtures have been evaluated with LCA study.

2. Methodology

2.1. Materials

The raw materials considered in this study included: (i) raw clay from a quarry in Renče in western Slovenia, which is used for brick manufacturing by local brick factory Goriške opekarne, and (ii) river sediment from Lake Ptuj, which is a reservoir for a local power plant on the Drava River in eastern Slovenia and is operated by Dravske elektrarne Maribor (DEM). The sediment from Lake Ptuj is regularly dredged and in-situ disposed on reservoir banks. Two different sediment samples were considered in this study, i.e. (i) sediment that has been naturally dehydrated for one year (Sed1) and (ii) sediment that has been naturally dehydrated for more than 10 years (Sed10). Sediment samples were stored in plastic containers in order to preserve the natural hydration during the testing period. The clay-sediment mixtures were obtained by mixing clay and river sediment in different ratios, with 8 different mixtures being considered within the study (Table 1).

The chemical composition of the raw materials was determined with X-ray fluorescence (XRF) technique using Thermo Scientific ARL Perform'X Sequential XRF. The XRF analysis was done on samples that were dried at 60 °C for 48 h and grinded below 125 μm with the vibrating disk mill (Siebtechnik TS250). The identification of the mineralogical phases of the raw materials was determined with X-ray diffraction (XRD) measurements using PANalytical Empyrean diffractometer with CuKα radiation and a nickel filter. The dried samples (grinded below 63 μm) were manually back-loaded into a circular sample holder with a 27 mm diameter in order to mitigate the possible preferred orientation. The samples were measured at the voltage of 45 kV, current of 40 mA and in the range of 4–70 °2θ.

Table 1
The material mixtures.

Mixture	Material
G0	clay
G1	clay and 10 wt% of Sed10
G2	clay and 20 wt% of Sed10
G3	clay and 30 wt% of Sed10
G5	clay and 10 wt% Sed1
G6	clay and 20 wt% Sed1
G7	clay and 30 wt% Sed1
G8	clay and 50 wt% Sed1

The loss on ignition was determined according to the standard EN 196–2 [53]. The ignited samples were mixed with Fluxana FX-X50-2 and melted into discs. In order to avoid the gluing of the melted disk to the platinum vessel, lithium bromide was added to the mixture of the ignited sample and Fluxana FX-X50-2. The particle size distribution was measured using Microtrac SYNC paricle-size analyzer (Model 5001, wet operation). The microstructure was analyzed using a scanning electron microscope (SEM) (JSM-IT500LV, Jeol, Tokyo, Japan) equipped with an energy dispersive X-ray analyser (EDXS, Link Pentafet, Oxford Instruments). The pore distribution of fired and unfired bricks was measured using Mercury Intrusion Porosimetry (MIP). Small representative fragments were dried to a constant mass in an oven (Binder) at 105 °C under laboratory atmosphere and analyzed using a Micromeritics®Autopore IV 9500 (Micromeritics, Norcross, GA, USA). Thermogravimetric analysis (TG/DTG) of the main raw materials were performed by Q5000IR analyser (TA instruments), on about 20 mg of each sample (G0 = 16.9790 mg, Sed1 = 26.2970 mg, G_{ind} = 12.5510 mg). Samples were placed in Pt crucibles and examined over the range from 24 to 960 °C using a heating rate of 10 °C/min in a synthetic air flow (25 ml/min).

2.2. Laboratory testing

The test samples for the ceramic-technological tests and firing in gradient kiln were shaped using the extruder at a vacuum of 0.82–0.85 kg/cm^2. For the ceramic-technological tests, the following shapes were analyzed: cylinders (mold diameter = 56.3 mm; extruded length = 55 mm), prisms (mold dimensions: 54.3 mm × 27.7 mm; extruded length = 150 mm) and tiles (mold dimensions: 57.2 mm × 9.3 mm; extruded length = 150 mm). Test samples with dimensions of 50 × 20 × 8 mm were prepared for firing in a gradient kiln.

The prepared test specimens were dried for 7 days at ambient room conditions, which was followed by drying in a dryer for 24 h at 60 °C and 8 h at 100 °C. The dried samples were then fired using heating rates of 150 °C/h. The maximum firing temperature reached up to 950 °C, with the dwelling time at maximum temperature being 2 h. The firing temperature was selected based on the data obtained by TG/DTG (Fig. 4), results from firing in gradient kiln (Fig. 5), and based on the data up to which temperature samples are fired at the producer plant. Namely, up to 950 °C clay minerals and carbonates decomposed, while the vitrification does not yet take place [54]. Finally, the samples were cooled inside the furnace. The determination of linear shrinkage, water absorption, compressive and flexural strength of the fired samples was conducted according to the requirements specified in the EN 772 standard series [1,55–59].

2.3. Industrial test

An industrial test, i.e. a full-scale test was conducted in an industrial setting at the Goriške opekarne brick production plant, using actual industrial-sized equipment under real production conditions. The goal was to demonstrate that the process or technology can be successfully implemented on a real scale. For this purposes 20 tons of the Drava River sediment sample Sed1 was used. The test was performed with the addition of 20 wt% of sediment (dry mass) in the brick-making material mixture. First, 13.2 vol % of raw clay and 26 vol % (i.e. 20 wt%) of Sed1 sample were mixed and discharged into a feed-shut, where clay and sediment mixture were further mixed with already pre-mixed 60.8 vol % of marl. After mixing of the raw materials, the brick-making mixture was carried by a conveyor-belt to the grinder, where the mixture was grinded and fed to the vacuum extruder.

Next, the test bricks were formed by wire cutting of the extruded mass. The formed bricks were then dried for a period of 40 h in a tunnel drier, with drying temperatures ranging from initial temperature of 37 °C to the final temperature of 90 °C. Finally, the test bricks were fired in a tunnel kiln for a period of 25 h, with maximum temperature in the kiln reaching up to 880 °C. The full drying and firing program can be seen in the supplementary data (Appendix A). The quantity of the so produced bricks was app. 10.000 units. The final testing of the bricks produced during the pilot production (e.g. water absorption, compressive strength etc.) was conducted according to the requirements specified in the EN 772 standard series [1,55–59].

2.4. LCA analysis

2.4.1. Goal and scope of the study

The aim of the LCA study was to analyze the environmental impacts associated with the industrial production of bricks made from two different brick-making mixtures, i.e. original or traditional brick-making mixture (TB) and mixture of river sediment and traditional brick-making material (TB-SED). The main goal of the LCA study was to evaluate the potential environmental benefits of brick production where river sediment is used to replace a portion of clay in the brick-making mixture.

This LCA study has focused on the production stage of the brick life cycle, i.e. the so-called "cradle-to-gate" approach [60]. Hence, the LCA study has considered.

- the extraction and processing of raw materials and packaging,
- processing of river sediment,
- consumption of energy and water within the production process and transport of all materials to the brick production plant.

The schematic representation of the system boundaries is presented in Fig. 1.

The LCA study has also considered the consequential modelling principle (i.e. system expansion where necessary), which means that any potential environmental burdens of alternative or substitute materials (e.g. river sediment) are not transferred into the model [61]. By applying the system expansion (i.e. consequential modelling), the avoided environmental impacts of the in-situ sediment disposal have also been included within the system boundaries. The avoided in-situ disposal has been treated as the environmental credit, as it leads to reduction in the emissions of heavy metals to the environment (i.e. water and soil).

2.4.2. Life cycle inventory (LCI)

The production and processing of raw and auxiliary materials, the production and supply of energy and water, and the transport processes have been evaluated based on the LCI data given in GaBi Professional and Ecoinvent 3.8 databases. The transport processes take into account only the emissions and burdens associated with the operation of the transportation vehicles [5]. Two-way transport distances have been considered, which means that transport vehicles are fully loaded in the incoming direction and empty in the outgoing direction. The full life cycle inventory can be seen in the supplementary data (Appendix B).

The sediment from the Drava River is contaminated with heavy metals. However, the concentrations are still in the range that allows for a direct use of the sediment for the production of building materials without the need for pre-treatment of sediment [62]. Therefore, the river sediment used for brick production can be treated as a secondary material and thus modelled according to the Ecoinvent cut-off approach [63]. This means that there are no environmental burdens associated with the river sediment itself, but only burdens related to the processing of the sediment for further use in the brick production.

The processing of the sediment includes pumping of the dry sediment and subsequent material handling with the excavator. The data on energy consumption during the sediment pumping has been provided by DEM, with 0.466 l of diesel fuel being consumed for the pumping of 1 ton of dry sediment. The excavator operation (i.e. fuel consumption and related emissions) has been evaluated based on the LCI data given in GaBi Professional database. The environmental benefit due to the avoided in-situ disposal has been evaluated based on the results of leaching tests [62].

The considered traditional brick-making material and river sediment both contain a certain amount of organic matter (i.e. total organic carbon or TOC). The organic matter decomposes when the material is heated, with different decomposition stages occurring at different temperatures [62]. The mass loss can be determined with the loss on ignition (LOI) test, which defines the percentage of the loss in mass due to the decomposition of organic matter and release of volatile substances. By calculating the difference between the LOI percentage at 950 °C and LOI percentage at 550 °C, we can assess the amount of the CO_2 that is released due to the decomposition of the carbonates. It has been calculated that the share of carbonates in the traditional brick-making material is 3.8%, while the share of carbonates in the river sediment is 8.6%. This means that more CO_2 is released during the firing of brick-making material mixed with sediment when compared to the firing of traditional brick-making material.

2.4.3. Life cycle impact assessment (LCIA)

The environmental impacts have been evaluated with CML 2001 (version Aug. 2016) life cycle impact assessment (LCIA) method. CML 2001 is an LCIA method that restricts quantitative modelling to early stages in the cause-effect chain to limit uncertainties, with

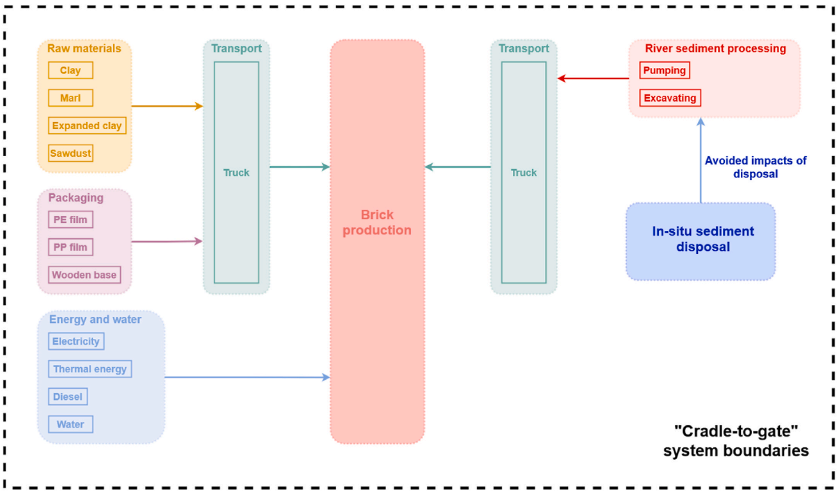

Fig. 1. The schematic representation of the system boundaries.

results being grouped in midpoint categories according to commonly accepted groupings (e.g. climate change) [64]. The main principles behind the CML 2001 impact assessment methodology are based on ISO 14040 and 14044 standards, with CML 2001 being one of the most commonly used impact assessment methods when evaluating life cycle of bricks [40].

The results of the CML 2001 LCIA method have been presented in terms of the following impact potentials: Abiotic Depletion Potential for non-fossil resources (ADP el., unit: kg Sb eq.), Abiotic depletion potential for fossil resources (ADP fos., unit: MJ), Acidification Potential (AP, unit: kg SO_2 eq.), Eutrophication Potential (EP, unit: kg $PO_{4\text{-}3}$ eq.), Global Warming Potential (GWP, unit: kg CO_2 eq.), Human Toxicity Potential (HTP, unit: kg DCB eq.), Marine Aquatic Ecotoxicity Potential (MAETP, unit: kg DCB eq.), Ozone Depletion Potential (ODP, unit: kg R11 eq.), Photochemical Ozone Creation Potential (POCP, unit: kg Ethene eq.) and Terrestric Ecotoxicity Potential (TETP, unit: kg DCB eq.).

3. Results and discussion

3.1. Physicochemical characteristics of raw materials

The mineralogical analysis of the raw materials is presented in Fig. 2, while the chemical composition of the analyzed raw materials is summarized in Table 2. The raw materials are mainly composed of quartz, with illite/muscovite, chlorite/kaolinite, feldspars, calcite and zeolites also being presented in different proportions. The main Fe-bearing minerals are muscovite [65] and chlorite [66]. Although hematite was not undoubtedly confirmed by XRD, it is most probably also presented in amounts bellow XRD detection limit (< approx. 1 wt%), since it is responsible for the red colour of the bricks even though it is only present in small quantities [67]. The main difference between the clay and the two sediment samples was in the CaO and MgO contents, with the CaO and MgO contents being two to three times higher in sediments when compared to clay (Table 2). This is due to the dolomite presence in the two sediment samples; namely, the dolomite content was not present in the clay.

The mineralogical composition of the brick-making materials plays an important role in the brick manufacturing, as the processes of shaping, drying and firing of bricks depend more on the mineral composition than on the chemical composition of materials. The results presented in Fig. 2 and Table 2 indicate that the Drava River sediment has a similar mineralogical and chemical composition as the brick-making clay, which indicates that the sediment could be used as a substitute for clay in the production of bricks. According to historic mines Pb–Zn mines in the upstream area sediments of Drava river have increased Pb, Zn and Cd content [68]. However, the heavy metal concentrations determined in sediments still allow their use for the production of construction materials without the need for pre-treatment [62]. Moreover, in preliminary laboratory scale study the leaching tests confirmed that fired brick production process successfully immobilized the heavy metals, resulting in leached concentrations in compliance with current regulations [62].

The results of the particle size distribution analysis of the raw material are shown in Fig. 3. The considered clay included a high percentage of particles that are less than 2 μm in size, while the percentage of clay fraction was lower in both sediment samples. Fig. 3 also shows that all raw material samples are characterized by a bimodal particle size distribution, with size distribution fraction ranging from fine fractions (clay) to coarse fractions (sand). It can be also seen from Fig. 3 that the particle size distribution of the considered two sediment samples were generally similar, with smaller difference being for the fractions ranging between 10 and 100 μm.

TG/DTG analysis was performed on raw mixture with Sed1 from industrial bricks (G ind) as well as for the main constituents of laboratory scale raw mixture, namely clay G0 and sediment Sed1 (Fig. 4) in order to evaluate thermal stability and reaction kinetics of the materials. In all samples the main weight loss up to 200 °C corresponds to sharp peaks on DTG curve at around 60 and 120 °C (Fig. 4). Since clay G0 had the highest peak intensity the weight loss can be attributed to hygroscopic and physical bound water in clay minerals [69–71]. At higher temperatures, TG curves decline more gradually (Fig. 4). Small peak on DTG curve appears at around 260 °C, evident only in clay sample (G0) and probably indicating the final stage of clay dehydration [69]. In sediment a broad peak was detected on DTG curve in temperature range between 220 and 420 °C, which can be attributed to the initial stage of illite/mus-

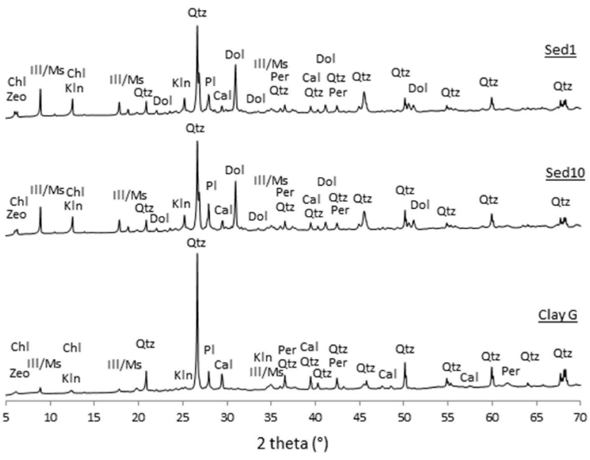

Fig. 2. XRD patters of raw materials (Qtz = quartz, Dol = dolomite, Pl = plagioclase, Ill/Ms = illite/muscovite, Kln = kaolinite, Chl = chlorite; Cal = calcite, Zeo = zeolite, Per = periclase).

Table 2
Chemical analysis of raw materials.

Composition (wt.%)	Clay	Sed10	Sed1
Na_2O	0.81	1.35	1.23
MgO	1.73	4.90	5.33
Al_2O_3	15.59	15.44	15.78
SiO_2	58.21	48.01	44.87
P_2O_5	0.10	0.19	0.24
SO_3	0.17	0.41	0.49
K_2O	2.25	2.23	2.30
CaO	3.64	6.15	7.12
TiO_2	0.71	0.81	0.83
V_2O_5	0.02	0.02	0.03
Cr_2O_3	0.02	0.02	0.01
MnO	0.21	0.09	0.08
Fe_2O_3	6.05	5.36	5.49
Co_3O_4	0.00	b.d.l.	b.d.l.
NiO	0.01	0.00	b.d.l.
CuO	0.00	0.00	b.d.l.
ZnO	0.01	0.13	0.16
As_2O_3	0.08	0.07	0.09
Rb_2O	0.02	0.02	0.02
SrO	0.02	0.02	0.02
ZrO_2	0.03	0.03	0.03
BaO	0.04	0.09	0.12
La_2O_3	b.d.l.	0.01	0.01
PbO	b.d.l.	0.02	b.d.l.
LOI 550 °C	6.48	7.04	7.21
LOI 950 °C	10.25	14.63	15.79

b.d.l = below detectable level.

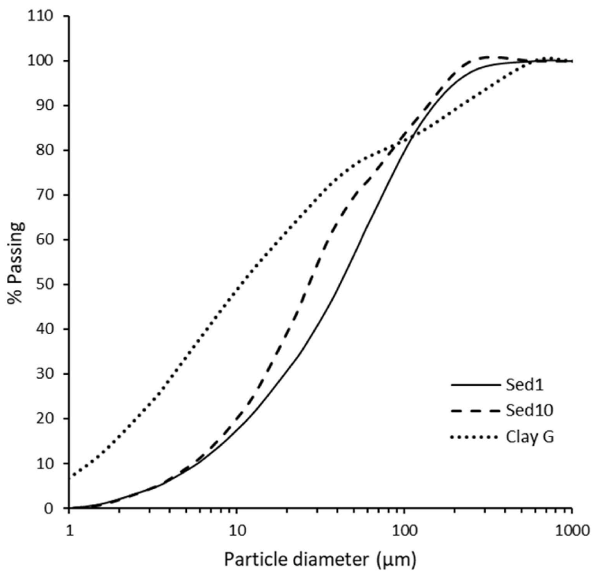

Fig. 3. The particle size distribution of raw materials.

covite dehydroxylation [69]. At higher temperature a small peak is evident at around 520 °C, most likely indicating kaolinite dehydroxylation [72]. Above 620 °C TG curve show sudden simultaneous declination in all samples, which can be attributed to the decomposition of carbonates [69]. However, DTG curve shows that peak intensity and upper temperature limit increases by elevated sediment content (Fig. 4). Wider peak of carbonate decomposition is probably related to increased illite/muscovite content in sediment and the final break down of illite structure, which takes place between 700 and 850 °C [72]. According to TG results between 24 and 960 °C minimum weight loss was measured in clay G0 (8.5%), intermediate weight loss in industrial raw mixture (10.1%) while maximum weight loss appeared in sediment Sed1 (14.7%).

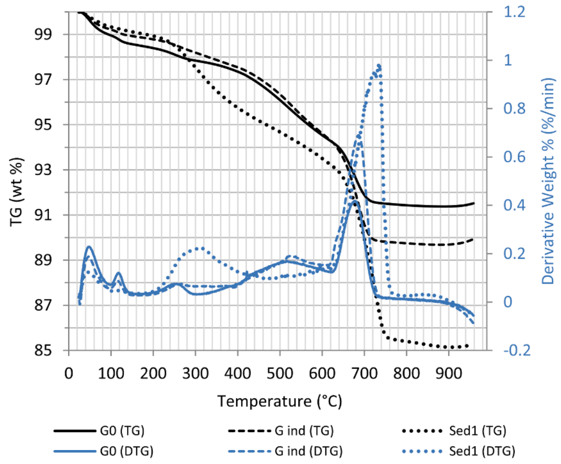

Fig. 4. Thermal characterization of brick raw materials (G0 = clay, used for laboratory experiments; G ind = raw mixture of industrial bricks; Sed1 = river sediment, naturally dehydrated for 1 year).

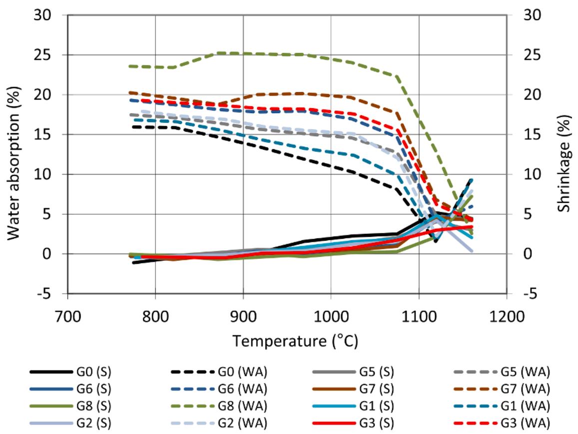

Fig. 5. The effect of the firing temperature on shrinkage and water absorption of the laboratory brick samples (S denotes shrinkage and WA water absorption).

3.2. Physical and mechanical properties of laboratory brick samples

The effect of the firing temperature on shrinkage and water absorption of the laboratory samples can be seen in Fig. 5. The tabular representation of the results is provided in the supplementary data (Appendix C). The substitution of a portion of clay in the brick-making mixture with the river sediment led to a reduced firing shrinkage and significantly increased open porosity. For example, the shrinkage and the water absorption of the pure clay sample (sample G0) at 950 °C was 1.6% and 11.9%, while the shrinkage and the water absorption of the sample with clay and 50 wt% of sediment (sample G8) at 950 °C was −0.3% and 25%. It can be also seen from Fig. 5 that the sediment age generally did not have a significant impact on the fire shrinkage and water absorption rates. There was a minimal difference between samples with 1-year old sediment and 10-year old sediment in terms of firing shrinkage, while the water absorption is in general about 15% higher for samples with 1-year old sediment.

The main parameters related to the porosity (MIP) of unfired laboratory clay samples dried to constant mass at 105 °C and samples fired at 950 °C are presented in Table 3. The porosity of fired samples was higher than the porosity of unfired samples, with the lowest porosity being detected for both unfired and fired pure clay samples (G0). Furthermore, the porosity increases with the addition of the river sediment. In addition, the porosity was in negative correlation to the bulk density due to the material compaction. Pores exhibited unimodal distribution in all samples, with fired samples having more smooth curves than unfired samples (Fig. 6). All in all, it was observed that the pore sizes increase with the addition of increasing amounts of the river sediment, which can be correlated to increased carbonate content, which is introduced into brick raw mixture by sediment addition (Table 2, Fig. 2). Namely, when carbonates are presented in brick-making raw mixture their thermal decomposition causes formation of fissures and pores under 1 μm in size, which results in increased porosity [54]. Similar, higher carbonate content in bricks containing 1-year deposited sediment resulted in higher porosity than when over 10-years deposited sediment with lower carbonate content was used [54]. Further, increased porosity led to about 15% higher water absorption of samples with 1-year old sediment comparing to over 10-years deposited sediment (Fig. 5).

Fig. 7 shows the results of SEM analysis, which was performed to evaluate the morphology and microstructure of the selected laboratory bricks samples. Electron microscopy revealed elongated pores with individual angular to sub-angular shaped pores was

Table 3
The results of the Mercury intrusion porosity analysis for the laboratory brick samples.

Mixture	Firing Temp (°C)	Porosity (%)	Average pore diameter (μm)	Median pore diameter (μm)	Bulk density (g/ml)
G0	–	23.08	0.03	0.08	2.03
G5	–	23.78	0.03	0.14	1.96
G6	–	25.56	0.04	0.16	1.94
G7	–	27.15	0.06	0.23	1.87
G0	950	27.46	0.38	0.63	2.01
G1	950	29.25	0.34	0.62	1.89
G2	950	30.88	0.35	0.66	1.85
G3	950	33.04	0.32	0.78	1.81
G5	950	29.82	0.36	0.74	1.73
G6	950	32.80	0.32	0.77	1.81
G7	950	34.52	0.39	0.82	1.76

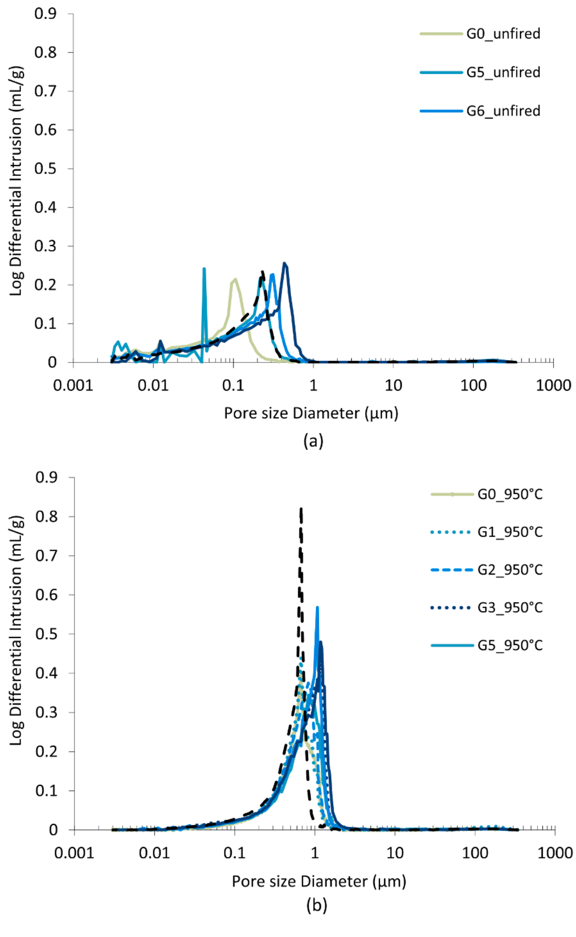

Fig. 6. Log differential intrusion vs. pore size (MIP) of the unfired samples (a) and samples fired at 950 °C (b).

mainly around 10–50 μm (Fig. 7), which might be attributed to burnt organic particles, derived from river sediments [62]. However, pores larger than 2 μm were not detected by MIP, most likely because MIP can only detected the entry size of spherical pores. In addition, all samples had matrix-grain-supported to matrix-supported texture, where minerals are surrounded by the so called "binding matrix" [73]. The matrix is crystalline with poor vitrification, which is characteristic for clay bricks with carbonate content [54].

EDXS elemental maps of Si, Ca and Mg-bearing mineral phases show that the majority of grains are quartz, with no clear differences in the quantity between the samples (Fig. 7). The presence of calcium rich grains could be attributed to the plagioclase content in the raw materials (Fig. 2). However, it should be noted that among the Ca-bearing phases, gehlenite and anorthite could be the newly formed phases during the sintering process [67,74]. The smallest amount of Ca-rich grains were presented in the pure clay sample, which can be attributed to the low content of Ca due to the absence of dolomite (Fig. 2). Also, clay had a much lower Mg content compared to the river sediment (Table 1). Therefore, the Mg-rich grains were observed only in the samples with the addition of the river sediment, which was probably due to the remnants of dolomite after calcination [54].

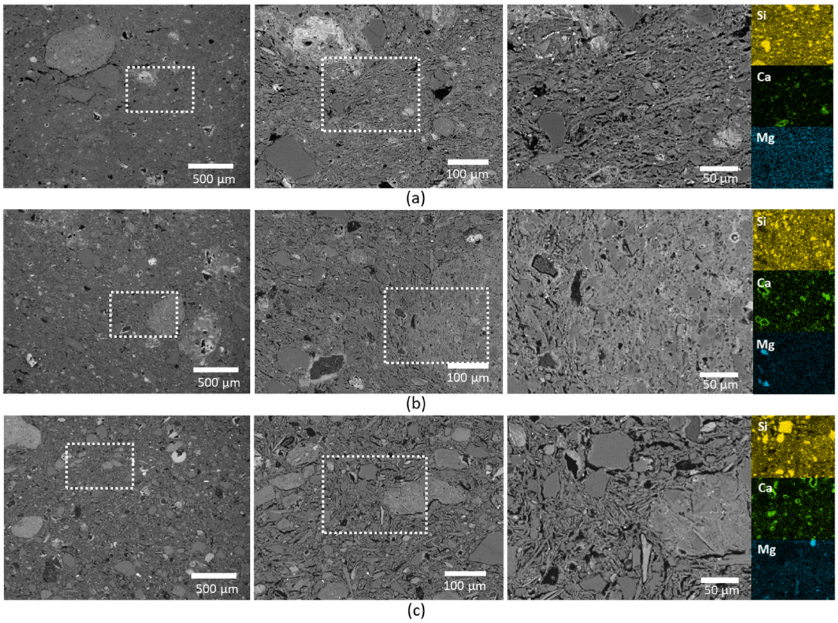

Fig. 7. SEM images and EDS elemental maps of Si, Ca and Mg for the selected laboratory brick samples fired at 950 °C, where (a) is pure clay, (b) is clay with 10 wt% of river sediment and (c) is clay with 30 wt% of river sediment.

The physical and mechanical properties of the fired laboratory brick samples are presented in Table 4. The addition of river sediment did not have a significant impact on the drying shrinkage, while the firing shrinkage decreased with the addition of increasing amounts of the river sediment (Table 4). Since reversible phase transition of α into β quartz is accompanied by notable expansion [75] but since clay G0, which showed the largest firing shrinkage, contains maximum amount of SiO_2, it is not likely that phase transition of α into β quartz is responsible for the observed decrease in firing shrinkage (Table 2, Table 4). Because the firing shrinkage decreased with the addition of increasing amounts of the river sediment, it is most likely that reduced firing shrinkage is related to increased carbonate content and related growth of metastable phases during brick firing [67]. One of such metastable phases is free lime. In the presence of water it transforms into portlandite, which can further react with CO_2 to form calcite, resulting in volume increase [54]. Furthermore, the addition of increasing amounts of the river sediment resulted in decreased density and increased water absorption, which in turn led to a decrease in both bending and compressive strengths (Table 4). For example, the compressive strength of the pure clay sample (G0) was 79.5 MPa, while the compressive strength of the sample with the 50 wt% of sediment was 26.2 MPa. Nonetheless, the compressive strengths of the bricks made with river sediment were still above the limit value of 10 MPa specified in the standard EN 772–1 [1].

3.3. *Industrial pilot production*

The characteristics of bricks made during the full-scale industrial test are summarized in Table 5. The addition of 20 wt% of river sediment to the brick-making material led to an increased plasticity (from 21.7% for original mixture to 22.6% for mixture with river sediment) and $CaCO_3$ content in total mass (from 6.6% for original mixture to 10.2% for mixture with river sediment). This increase is the result of the finer granulometric composition and the higher total organic content of the river sediment. Therefore, the addition of 20 wt% of river sediment to the brick-making material had an impact on the water-mineral binding due to the different physical and chemical characteristics of the river sediment when compared to clay [54].

The addition of the river sediment to the brick-making mixture did not have an impact on the raw brick density, while there was a slight difference in the weight loss after 40 h of drying (Table 5). The drying weight loss of the original mixture was between 17.5%

Table 4
Physical and mechanical properties of the laboratory samples.

Mixture	Drying shrinkage by length (%)	Firing shrinkage by length (%)	Density (g/cm^3)	Water absorption (%)	Bending strength (MPa)	Compressive strength (MPa)
G0	8.5	0.3	1.85	10.9	15.6	79.5
G1	7.6	0.2	1.84	13.0	13.8	68.8
G2	7.5	0.0	1.77	15.0	12.5	48.1
G3	7.6	−0.1	1.74	17.3	11.1	50.0
G5	8.5	0.0	1.78	14.5	12.1	73.9
G6	8.7	−0.1	1.73	16.8	11.3	54.7
G7	8.1	−0.2	1.68	19.1	10.0	52.8
G8	8.4	−0.3	1.55	24.2	7.0	26.2

Table 5
The characteristics of bricks made during the industrial test.

Parameter	Original mixture	Mixture with the addition of 20 wt% of river sediment
$CaCO_3$ content in total mass [%]	6.6 ± 0.3	10.2 ± 0.3
Pfefferkorn plasticity [%]	21.7 ± 0.8	22.6 ± 0.9
Raw bricks density (kg/m^3)	2050 ± 50	2057.5 ± 35.5
Burned brick density (kg/m^3)	1970 ± 10	1885 ± 79
Drying shrinkage (%)	5.25 ± 0.25	4.7 ± 0.1
Weight loss during drying (%)	18.0 ± 0.5	17.15 ± 0.15
Burning shrinkage (%)	5.25 ± 0.25	4.7 ± 0.1
Weight loss during burning (%)	6.25 ± 0.25	9.1 ± 0.1
Water absorption (%)	10.5 ± 0.5	15.25 ± 0.15
Compressive strength (MPa)	38 ± 1	25.85 ± 1.15

and 18.5%, while the averaged drying weight loss of the mixture with the river sediment was 17.1%. The drying shrinkage of the bricks made with the original mixture was between 5% and 5.5%, while the average shrinkage of the bricks with the addition of the river sediment was 4.7% (Table 5). The higher value of the shrinkage during the drying generally results in higher stresses in the brick, which can lead to the breaking of dry products. The addition of the river sediment has reduced the drying shrinkage, but it has also resulted in the increase in the fracture percentage. This was probably due to the accelerated shrinkage during the drying process, where the final shrinkage value is lowered.

It can be further seen from Table 5 that the addition of the river sediment to the brick-making mixture resulted in the higher weight loss during the burning, which increased form 6.25% (original mixture) to 9.1% (mixture with the addition of the river sediment). A part of the weight loss can be attributed to the crystal bound water and $CaCO_3$/$MgCO_3$ decomposition [54]. In addition, the increased burning weight loss of the bricks made with the addition of the river sediment is due to the higher content of organic impurities in the sediment. The decomposition of higher content of organic matter during the burning of bricks with the addition of the river sediment leads not only to higher mass loss, but also to formation of white spots on the surface of bricks, which can be seen on Fig. 8.

The addition of the river sediment to the brick-making mixture resulted also in higher water absorption coefficient, which increased from 10.5% for bricks made with the original brick-making mixture to 15.25% for bricks made with the addition of the river sediment. The shrinkage of the bricks with added sediment was lowered during sintering, while the mass remained similar to the bricks from the regular production. Consequently, the porosity increased with the addition of the river sediment, which had a direct impact on the water-absorbing properties.

Finally, the bricks made with the addition of the river sediment had lower compressive strength, with the average compressive strength being 32% lower when compared to the bricks from the regular production (Table 5). The lower compressive strengths of the bricks made with the addition of the river sediment was primarily due to the higher mass loss and increased porosity, which affect the weakening of the structural strength. Nonetheless, the average compressive strength of the bricks made with the addition of the river sediment was still above the limit value of 15 MPa specified in the standard EN 772–1 [1].

3.4. LCA analysis

Fig. 9 shows the relative contributions of raw and auxiliary materials, supporting processes, transport and energy and water requirements to the environmental footprint of the production of bricks made from the traditional brick-making material (i.e. clay and marl). Brick firing (i.e. gas induced thermal energy) and brick-making raw material (i.e. clay and marl) are responsible for the majority of the total environmental burden (Fig. 9). For example, the thermal energy and raw material requirements represent

Fig. 8. The formation of white spots due to the of loss of the organic phase during the burning of the bricks made with the addition of 20 wt% of river sediment.

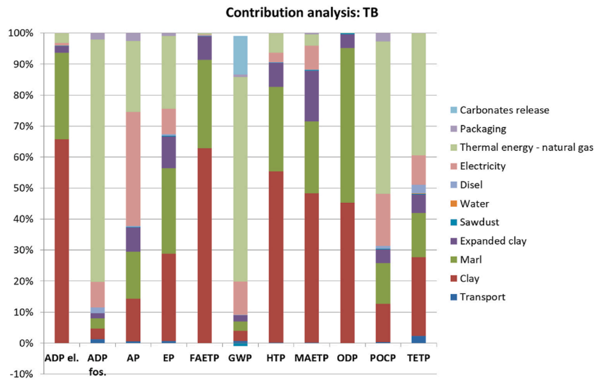

Fig. 9. The contributions of different materials and processes to overall environmental impact associated with the production of bricks made from the traditional brick-making material (i.e. clay and marl).

more than 75% of the total impact in terms of 10 out of 11 considered impact categories, with the exception being the impact in terms of AP where they represent 52% of the total impact. All other materials and processes generally have much smaller environmental impact, or they contribute more significantly only in terms of an individual impact category, such as electricity requirements in terms of AP (37%).

Fig. 10 shows the relative contributions of raw and auxiliary materials, supporting processes, transport and energy and water requirements to the environmental footprint of the production of bricks made from the mixture of river sediment and traditional brick-making material. In general, the relative contributions are similar to the results shown in Fig. 9. The main difference is the greater contribution of transport and carbonates release in terms of GWP, with transport representing 9% and carbonates release 18% of the total impact in terms of GWP. However, this was expected due to the longer transport distances (mainly as a result of sediment transport to the production site) and higher percentage of organic matter in the river sediment.

It can be also seen from Fig. 10 that transport has a positive impact in terms of photochemical ozone creation potential (POCP). The photochemical ozone is generated by sunlight-initiated oxidation of volatile organic compounds (VOC) and carbon monoxide in the presence of nitrogen oxides (NOx) [76]. The reaction of VOC with different oxidants (O_3, NO_2, etc.) can have either positive or negative impact on the photochemical ozone creation. Thus, the negative value of the transport in terms of the POCP is related to the separation of the NOx emissions in the NO_2 and NO emissions. Namely, reaction of NO and O_3 to NO_2 and O_2 during the night time leads to the positive impact (i.e. reduction) in terms of POCP [5].

The comparison between the environmental performance of brick production from traditional brick making material and mixture of sediment and traditional brick-making material is shown in Fig. 11. Production of bricks made form mixture of river sediment, clay and marl leads to reduction in environmental footprint in terms of all impact categories except in terms of GWP (Fig. 11). In terms of GWP, the production of bricks made from mixture of river sediment and traditional brick-making material leads to 16% increase in CO_2 emissions. As mentioned, this is a result of longer transport distances (i.e. higher consumption of diesel fuel) and larger quantities of released CO_2 during the firing of the bricks where the portion of clay is replaced with river sediment.

The CO_2 emissions could be decreased by considering a production location closer to the sediment source. For example, there is a Wienberger's brick production facility in Ormož, which is located 23 km east of Lake Ptuj (i.e. sediment reservoir). The production in Ormož would lead to a 90% decrease in the sediment transport distance when compared to production in Renče, which is located 250 km from the Lake Ptuj. Hence, the impact in terms of GWP would decrease for nearly 10%, as it can be seen from Fig. 11. However, this is also the maximal reduction in CO_2 that could be achieved without changing the technological process etc., since the po-

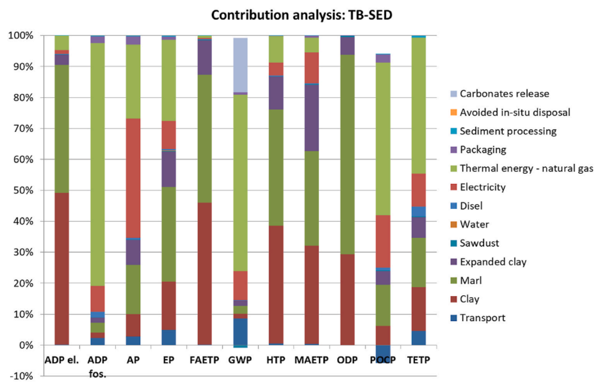

Fig. 10. The contributions of different materials and processes to overall environmental impact associated with the production of bricks made from the mixture of river sediment and traditional brick-making material.

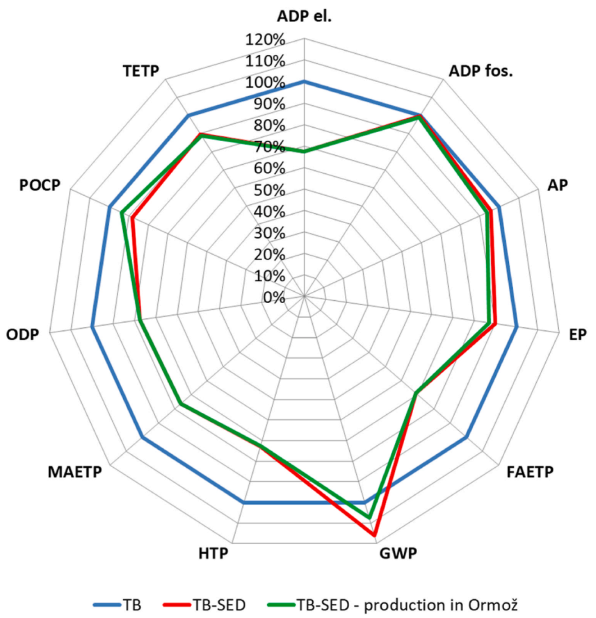

Fig. 11. The comparison of the environmental performance of different brick-making materials and the impact of an alternative production location for bricks with the addition of river sediment.

tential production location in Ormož is as close to the source of river sediment as possible. Therefore, any further potential reduction in CO_2 would have to be based either on modification of the production process or pre-treatment of sediment.

4. Conclusions

In this study, we have investigated the potential use of the untreated Drava River sediment as a substitute material for clay in the production of fired bricks. The optimal sediment to clay ratio has been determined based on the laboratory tests, with the selected brick-making mixture then being used for brick production at the industrial scale (pilot production). The following concluding can be drawn based on the study results.

- The Drava River sediment has a similar mineralogical and chemical composition as the considered clay, with the main difference being in MgO and CaO contents due to the lack of dolomite in clay. Hence, the Drava River sediment appears as a suitable candidate to substitute clay in brick-making material.
- Increased carbonate content in Drava river sediment results in higher weight loss during firing at temperatures up to 960 °C, comparing to firing pure brick-making clay.
- The replacement of a portion of clay with river sediment did not have a significant impact on the drying shrinkage, while the firing shrinkage decreased with the addition of increasing amounts of the river sediment. The expansion is most probably related to the growth of metastable phases in presence of carbonates during brick firing.
- The compressive strength of the laboratory specimens (fired at 950 °C) decreased with the addition of increasing amounts of the river sediment, with compressive strength dropping from 73.9 MPa (10 wt % of sediment) to 26.2 MPa (50 wt % of sediment). The density and porosity had the greatest impact on the mechanical properties, with the bulk density decreasing and water absorption increasing with the addition of increasing amounts of the river sediment.
- The compressive strength of the hollow bricks from the pilot production decreased from 38 MPa (original mixture) to 26 MPa (mixture with river sediment). However, the compressive strength of the bricks made with the river sediment was still above the limit of 10 MPa specified in the relevant regulatory standard.
- The results of the LCA has shown that the industrial production of bricks where a portion of clay is replaced with river sediment in the brick-making mixture (i.e. 20 wt % of sediment) leads to an overall decrease in environmental footprint, particularly in terms of lower toxicity impact and reduced raw material consumption. However, there in an increase in CO_2 emissions due to the higher percentage of organic matter in the river sediment, which is released when carbonates start decomposing during the firing of bricks.

CRediT authorship contribution statement

Mojca Božič: Writing – review & editing, Data curation. Lea Žibret: Writing, Investigation, Formal analysis, Data curation. Davor Kvočka: Software, Investigation, Validation, Formal analysis, Data curation, Writing – review & editing. Alenka Mauko Pranjić: Formal analysis, Writing – review & editing.Boštjan Gregorc: Writing – review & editing, Funding acquisition. Vilma Ducman: Conceptualization, Methodology, Supervision, Writing – review & editing.

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.jobe.2023.106470.

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