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Corresponding Author: Mrs. Rawan Khalil Mlih, ph.D

Corresponding Author's Institution: Juelich Forschungszentrum

First Author: Rawan Khalil Mlih, ph.D

Order of Authors: Rawan Khalil Mlih, ph.D; Franciszek Bydalek, Ph.D; Erwin Klumpp, Prof.; Nader Yaghi, Dr.; Roland Bol, Prof.; Jannis Wenk, Dr.

Abstract: Light expanded clay aggregates (LECA) have been increasingly used as substrate material for constructed wetlands given their phosphate removal capacity, mechanical strength, hydraulic conductivity and their plant rooting and biofilm growth supporting structure. This review summarizes the current literature on LECA-based constructed wetlands. Removal performances for main wastewater parameters phosphate, nitrogen species, suspended solids and oxygen demand are tabulated. Both, physical and biological water purification processes in LECA wetlands are discussed. Additional emphasis is on design and layout of LECA wetlands for different types of wastewater, under different climatic conditions and to improve treatment performance in general. LECA life cycle considerations include sourcing, production energy demand, reuse and recycling options for spent wetland substrates, for example as soil amendment. Research and development opportunities were identified for structural and compositional LECA modification to obtain tailored substrates for the use in water treatment and specific treatment tasks. Beyond traditional wastewater contaminants the fate of a wider range of contaminants, including organic trace contaminants, needs to be investigated as high Fe, Al and Ca oxides content of LECA substrates provide adsorptive sites that may facilitate further biological interactions of compounds that are otherwise hard to degrade.

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Prof. Dr. Wulf Amelung
Prof. Dr. Jan Vanderborght
Prof. Dr. Harry Vereecken

Forschungszentrum Jülich GmbH · IBG-3 · 52425 Jülich

Ihr Zeichen:
Ihre Nachricht vom:
Unser Zeichen:
Unsere Nachricht vom:

Ansprechpartner: Mrs. Rawan Mlih
Organisationseinheit; IBG-3

Tel.: 02461 61-6992
Fax: 02461 61-2518
r.bol@fz-juelich.de

August 28, 2019

Dear Dr. Jan Vymazal (Editor-in-Chief, Ecological Engineering)

Manuscript title: Light-expanded clay aggregate (LECA) as a substrate in constructed wetlands–A review

We are pleased to submit a new review manuscript for your consideration in Ecological Engineering Journal entitled 'Light-expanded clay aggregate (LECA) as a substrate in constructed wetlands–A review'. We, the authors believe that the topic fits aims and scope of the journal extremely well. The review manuscript presents has not been submitted or published elsewhere. All authors have approved the final version of the manuscript.

The review summarized the current knowledge of Light Expanded Clays Aggregates (LECA) as a substrate material in constructed wetlands (CWs). Research gaps are highlighted for example on crushed and structurally modified LECA. It also addressed the background mechanisms for LECA's performance as one of the best adsorbents for a wide range of pollutants in general and for P in particular due to its high Fe, Al, and Ca oxides contents. Special focus was given to the technical aspects and optimal design considerations for the incorporation of LECA in different types of CWs. Saturated LECA can in principle be used as a soil amendment and nutrient releasing fertilizer, thus contributing to full cycling of clay based products in the context of a more sustainable bio-economy. The review also explores the environmental conditions underlying the high production costs, high energy consumption and potential issues of recycling of the LECA material upon saturation.

We look forward to your comments and those of the peer-reviewers in due course.

Yours Sincerely,

Mrs. Rawan Mlih, Ph.D

Conflict of interest

Declarations of interest: **none.**

There's no financial or personal interest or belief that could affect this work objectivity

The authors

Highlights:

- Gap/strength analysis of LECA as a substrate material in constructed wetlands.
- High Fe, Al, and Ca oxides content underpin LECA pollutant removal and adsorbance.
- LECA has indirect effects on parameters controlling biochemical efficiency of CWs.
- Technical aspects and optimal design considerations for LECA in different CWs evaluated.
- Saturated LECA potential use as soil amendment and fertilizer reviewed.

Revised title page

Light-expanded clay aggregate (LECA) as a substrate in constructed wetlands–A review

Rawan Mlih[*1, 5], Franciszek Bydalek[2, 3], Erwin Klumpp[1], Nader Yaghi[4], Roland Bol[1], Jannis Wenk[3]

[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany

[2] GW4 NERC Centre for Doctoral Training in Freshwater Biosciences and Sustainability, Museum Avenue, Cardiff CF10 3AX, United Kingdom

[3] Department of Chemical Engineering, Water Innovation & Research Centre (WIRC), University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom

[4] Department of Chemistry, Faculty of Sciences, University of Hebron, 90100 Hebron, Palestine

[5] Institute for Environmental Research, Biology 5, RWTH Aachen University, Worringerweg 1, 52074 Aachen, Germany

*Corresponding author: Rawan Mlih, r.mlih@fz-juelich.de

Name	Affiliation	Email address
Rawan Mlih[*1, 5]	[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany [6] Institute for Environmental Research, Biology 5, RWTH Aachen University, Worringerweg 1, 52074 Aachen, Germany	r.mlih@fz-juelich.de
Franciszek Bydalek[2, 3]	[2] GW4 NERC Centre for Doctoral Training in Freshwater	fab52@bath.ac.uk

	Biosciences and Sustainability, Museum Avenue, Cardiff CF10 3AX, United Kingdom [3] Department of Chemical Engineering, Water Innovation & Research Centre (WIRC), University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom	
Erwin Klumpp[1]	[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany	e.klumpp@fz-juelich.de
Nader Yaghi[4]	Department of Chemistry, Faculty of Science and technology, Hebron University P.O BOX 40. Hebron, Palestine.	nadery@hebron.edu
Roland Bol[1]	[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany	r.bol@fz-juelich.com
Jannis Wenk[3]	[3] Department of Chemical Engineering, Water Innovation & Research Centre (WIRC), University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom	jhw46@bath.ac.uk

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Rawan Mlih[*1, 5], Franciszek Bydalek[2, 3], Erwin Klumpp[1], Nader Yaghi[4], Roland Bol[1], Jannis Wenk[3]

[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany

[2] GW4 NERC Centre for Doctoral Training in Freshwater Biosciences and Sustainability, Museum Avenue, Cardiff CF10 3AX, United Kingdom

[3] Department of Chemical Engineering, Water Innovation & Research Centre (WIRC), University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom

[4] Department of Chemistry, Faculty of Science and technology, Hebron University P.O BOX 40. Hebron, Palestine.

[5] Institute for Environmental Research, Biology 5, RWTH Aachen University, Worringerweg 1, 52074 Aachen, Germany

*Corresponding author: r.mlih@fz-juelich.de

Abstract

Light expanded clay aggregates (LECA) have been increasingly used as substrate material for constructed wetlands given their phosphate removal capacity, mechanical strength, hydraulic conductivity and their plant rooting and biofilm growth supporting structure. This review summarizes the current literature on LECA-based constructed wetlands. Removal performances for main wastewater parameters phosphate, nitrogen species, suspended solids and oxygen demand are tabulated. Both, physical and biological water purification processes in LECA wetlands are discussed. Additional emphasis is on design and layout of LECA wetlands for different types of wastewater, under different climatic conditions and to improve treatment performance in general. LECA life cycle considerations include sourcing, production energy demand, reuse and recycling options for spent wetland substrates, for example as soil amendment. Research and development opportunities were identified for structural and compositional LECA modification to obtain tailored substrates for the use in water treatment and specific treatment tasks. Beyond traditional wastewater contaminants the fate of a wider range of contaminants, including organic trace contaminants, needs to be investigated as high Fe, Al and Ca oxides content of LECA substrates provide adsorptive sites that may facilitate further biological interactions of compounds that are otherwise hard to degrade.

Keywords: Constructed wetlands; LECA; pollutants removal; phosphorous; nitrogen; adsorption.

1. Introduction

Traditional water treatment strategies employ a combination of physical, chemical and biological methods and require large investments in both infrastructure and operation (Goel 2006; Hendricks 2016). Nature-based solutions such as constructed wetlands (CWs) are considered as a viable alternative to conventional treatment systems. CWs are artificial wetlands for wastewater treatment, they consist of a flow-through substructure, saturated with water and are planted with adaptive vegetation (Verhoeven and Meuleman 1999). CWs can serve for a wide range of wastewater types at a comparable removal efficiency to conventional treatment, while requiring less investment costs, energy demand and maintenance (Vymazal 2010).

Similar to natural wetlands, CWs have been recognized for their multiple roles that combine environmental and societal benefits, including improving water quality, increasing water storage buffer capacity during draughts and storm events, restoring wildlife habitats and providing diverse recreational space within the urban landscape (Thorslund *et al.,* 2017). Since the introduction of the concept more than 50 years ago (Seidel 1961), the technology has advanced and CWs have been successfully used to treat an extensive range of domestic, agricultural and industrial wastewater streams under various climatic conditions (Calheiros *et al.,* 2007; Merlin *et al.,* 2002; Rozema *et al.,* 2016). CWs have been integral part of progressive ecological urban planning for example in the ‘sponge city concept’ and are well-established in decentralised water treatment schemes for smaller communities and rural settlements (Arheimer *et al.,* 2004; Liu *et al.,* 2017).

There are three major types of CWs (Wu *et al.,* 2015a): free surface water flow CWs, horizontal subsurface flow CWs and vertical subsurface flow CWs (Figure 1). Free surface water flow CWs closely replicate the natural cleaning processes occurring in natural wetlands and have been applied for different types of wastewater including those with high biological oxygen demand (BOD) and solids content (Ghermandi *et al.,* 2007; Vymazal 2013a). Both horizontal and vertical flow CWs are widely used (Luederitz *et al.,* 2001), while hybrid systems may combine advantages of each type of CW (Vymazal 2010). CWs range from simple, vegetated soil filtration beds to highly diverse multi-hectare systems that combine different types of CWs (Dunne *et al.,* 2012; Wu *et al.,* 2015a).

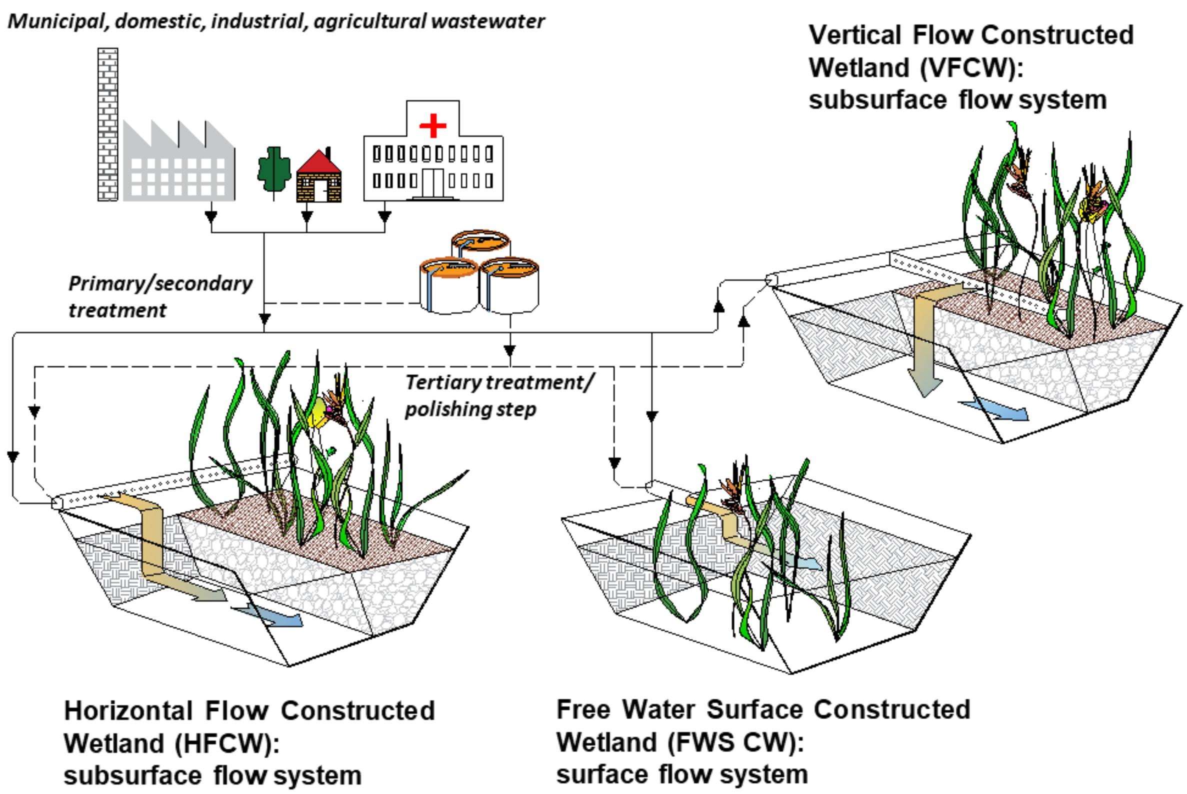

Figure 1 CWs are used for treatment of various types of wastewater, including rainwater, diluted municipal sewage and high strength industrial wastewater (i.e. effluents from slaughterhouses). CWs and can serve as either primary, secondary or polishing treatment step. There are three main types of constructed wetlands, classified based on the wastewater flow path: Free Water Surface CW (FWS CW), Horizontal Flow CW (HFCW) and Vertical Flow CW (VFCW).

The removal mechanism of pollutants in CWs is achieved through an integrated combination of biological, physical and chemical interactions among plants, the wetland substrates and microorganisms (Truu *et al.,* 2009; Vymazal 2005). Wetland substrate is a porous particulate packed bed filtration medium that creates the body of a CW. The substrate occupies the largest proportion of a CW and plays a central role in the purification process, including providing physical support for wetland plants (Wu *et al.,* 2015b). Biofilm forming microbial communities which drive biodegradation in CWs are influenced by physical and chemical properties of the substrate (Meng *et al.,* 2014). Substrate supplies adsorption sites for contaminants, facilitating various chemical processes taking place within substrate matrix (Calheiros *et al.,* 2009). Therefore, careful substrate selection is critical for optimized wetland performance. Location and depth of the substrate varies with the type of wetland. For vertical flow CWs the depth of the substrate ranges between 50-60 cm (Prochaska and Zouboulis 2009). The top 10-20 cm facilitate aerobic microbial activity and subsequent biodegradation, while the remaining 40-50 cm contribute to anaerobic processes including nitrogen removal, and phosphorus adsorption (Tietz *et al.,* 2007). The CW can consist of substrate layers of different granular sizes that increase towards the bottom drainage layer (Ávila *et al.,* 2015), and may include an additional organic substrate such as wood mulch (Myszograj and Bydałek 2016) or biochar (Zhou *et al.,* 2017). In the case of horizontal flow CWs, the effective substrate depth is between 25-60 cm (Carballeira *et al.,* 2017). Horizontal flow CWs can employ both mineral and organic substrate materials (Andreo-Martínez *et al.,* 2017), while multilayer structure is less common compared to vertical flow CWs. Free surface water flow CWs use 20-30 cm of rooting soil. For all types of CWs, small, round, evenly sized grains are most commonly used to fill the bed and average substrate diameters range from 3 to 32 mm (Tilley *et al.,* 2014).

Wetland substrates can be divided into natural and manufactured materials, which also include recycled and industrial by-products (Ballantine and Tanner 2010; Johansson

Westholm 2006; Wu *et al.,* 2015a). Natural substrates such as soil, sand, gravel and marine sediments have been traditionally used as filter materials in CWs. These substrates are widely available and require little pre-treatment prior to application (Healy *et al.,* 2007). However, clogging, poor adsorption capacity and low hydraulic conductivity are common problems associated with these substrates (Johansson Westholm 2006). More recently, both natural inorganic minerals such as anthracite, apatite, bauxite, calcite and zeolite (Molle *et al.,* 2011; Seo *et al.,* 2008; Stefanakis *et al.,* 2009) and organic materials such as biochar, rice straw, peat, and wood mulch became established as CW substrates (Gupta *et al.,* 2015; Kizito *et al.,* 2015; Xiong and Mahmood 2010). Recycled materials and by-products from mining, metal-making, construction, manufacturing and agriculture have also been used as substrate materials in CWs. Specifically alum sludge, coal and steel slag, fly ash, polyethylene plastic, oyster shells, tire chips, and construction waste such as bricks (Blanco *et al.,* 2016; Chyan *et al.,* 2013; Hu *et al.,* 2012; Shi *et al.,* 2017; Tatoulis *et al.,* 2017). Some substrates have been modified to improve treatment performance, mainly for better P removal (Ballantine and Tanner 2010; Johansson Westholm 2006), but also for improved ammonium (Zhang *et al.,* 2013) and heavy metal (Lian *et al.,* 2013) removal.

Recent reviews have compared the role of different substrates on the removal of nutrients, including P removal of both natural and manufactured substrates (Wang *et al.,* 2020) and summarizing the structural differences and inherent properties of unconventional substrates such as zeolite, rice husk, alum sludge among many others, and their capacity for substantial N and organics removal from wastewater under optimized operating conditions (Saeed and Sun 2012). Various substrates have been classified based on their ion-exchanging, P sorbing and electron donating properties (Yang *et al.,* 2018). In general, substrates rich in mineral oxides of calcium (Ca), aluminum (Al), and iron (Fe) such as limestone, biotite, muscovite, steel slag, and light weight expanded clays aggregates (LECA) have high capacities of P and N removal (Johansson Westholm 2006), while organic substrates such as rice straw, compost, and wood mulches can be utilized by microbes as electron donors and thus enhance nitrification and denitrification processes (Cao *et al.,* 2016). Specific studies have focused on substrates with extensive capacity for P removal such as clay bricks, fly ash, wollastonite, slag material, bauxite, shale, burnt oil shale, limestone, zeolite and LECA (Drizo *et al.,* 1999; Johansson Westholm 2006; Lima *et al.,* 2018).

This review focusses on suitability of LECA as a substrate in CWs and summarizes the current knowledge of LECA application in CWs design for wastewater treatment and its performance for a broad range of pollutants. The paper further examines the technical aspects of LECA incorporation into CWs design solutions with a wider attention to the importance and possibilities of LECA structural modifications enhancing the removal of different types of pollutants using CW technology. Moreover, the review aims to shed some light on the environmental concerns of LECA recycling and energy consumption.

2. Light Expanded Clay Aggregates (LECA)

2.1 Production, use and composition

LECA-like materials can be traced back to ancient Mediterranean civilizations (Chandra and Berntsson 2002). LECA is a subtype of light weight aggregates (LWA), a heterogeneous group of low-density materials used for various civil engineering and construction purposes (Al-Jabri *et al.,* 2005; Holm and Valsangkar 1993; Mladenovič *et al.,* 2004; Real *et al.,* 2016). LECA has been increasingly applied in storm water management schemes and urban green infrastructure including green roofs and walls, permeable pavements and thermal insulation concretes (Karami *et al.,* 2018; Molineux *et al.,* 2016; Pradhan *et al.,* 2018; Sailor and Hagos 2011; Sengul *et al.,* 2011). Commercial trademarks marketed worldwide include Filtralite®, Danish Leca®, LiaporTM, Stalite, Gravelite and Go Green (Baker *et al.,* 2014). The first use of LECA as CW substrate was reported in the early 1990s (Jenssen *et al.,* 1991). LECA is a strong but light aggregate with a water-resistant sintered ceramic matrix and a near-spherical shape (Cheeseman *et al.,* 2005). LECA has a water absorption capacity between 5-25% (Bogas *et al.,* 2012; Castro *et al.,* 2011; Nepomuceno *et al.,* 2018) and the cation exchange capacity of LECA is estimated by 9.5 cmol·kg^{-1} (Drizo *et al.,* 1999). Traditionally, clay minerals like montmorillonite or illite are used as a raw materials for LECA production (Nkansah *et al.,* 2012). Clay minerals are hydrous aluminium silicates with Fe, Mg and other alkaline and earth alkaline metals at variable amounts (Murray 2007). The chemical composition of LECA varies with the mineralogy of the raw clay material used (Shichi and Takagi 2000). More recently, a wider range of natural, artificial and recycled additives such as shale, apatite minerals, granite and marble mining residues, industrial by-products including fly ashes, wastewater sludge and contaminated soils have been incorporated to produce modified LECA (Ayati *et al.,* 2019; Cheeseman *et al.,* 2005; Molle *et al.,* 2011). LECA is manufactured by burning wet-formed clay granules at temperatures ranging from 1000-1300°C. Increased temperatures and burning times result in higher density and lower porosity product (Moreno-Maroto *et al.,* 2017). During burning the clay expands rapidly by gas generation through pore water evaporation, decomposition of carbonates and ferric oxides and combustion of intrinsic organic compounds and added expansion agents, including mineral oil, sawdust and chopped straw (Fakhfakh *et al.,* 2007; González-Corrochano *et al.,* 2009) . Due to chemical changes during burning, the final LECA product has a slightly different chemical composition than the raw material, lacking hydrated mineral forms and carbon. LECA composition is generally dominated by 5-6 major constituents; 60-70% SiO_2, 15-18% Al_2O_3, 4-7% Fe_2O_3, 1-4% MgO, CaO, Na_2O, other constituents contributing less than 1% (Table 1).

Table 1 The chemical composition of LECA produced from clay, marine clay, and fabricator sludge.

Reference	Sharifnia et al., 2016	Kalhori et al., 2013	Laursen et al., 2006	
LECA raw material	100% clay	100% clay	90% marine clay+ 10% semiconductor production sludge	
			a	b
SiO_2	61.67	64.83	70.7	69.2
Al_2O_3	18.51	15.05	15.3	15.6
Fe_2O_3	6.14	7.45	4.5	4.42
MgO	3.97	3.67	1.02	1.03
CaO	3.5	2.98	3.8	3.97
K_2O	3.28	2.55	1.39	1.5
Na_2O	1.54	1.1	0.51	0.54
TiO_2	0.65	0.63	0.57	0.6

SO_3	0.23	0.11	1.5	2.22
P_2O_5	0.19	0.13	nd	0.026
SrO	0.13	-	0.026	0.023
Cl^-	-	-	0.13	0.17
L.O.I	-	1.37	na	na
MnO	-	0.13	0.03	0.027
CuO	-	-	0.021	0.016
F	-	-	nd	0.21
ZnO	-	-	0.015	0.014
ZrO_2	-	-	0.101	0.053
BaO	-	-	0.36	0.31

LECA is available as granules (intact) or crushed (Figure 2), geotechnical and construction applications predominantly use granules, while crushed LECA is used in hydroponics and water filtration applications (Bahmanpour *et al.,* 2017). The LECA manufacturing process creates a pellet ranging from <1-32 mm with an average dry bulk density of about 400-600 kg m^{-3} and a smooth sintered ceramic outer shell that encloses the inner honeycomb structure (Ardakani and Yazdani 2014; Musa *et al.,* 2016). LECA manufacturing has not been optimized for applications that require a rather porous and sorbent surface as desired for CWs e.g. for P removal or as a matrix for biofilm growth. Crushing LECA creates almost twice the specific surface area and cation exchange capacity compared to uncrushed LECA (Kalhori *et al.,* 2013; Stevik *et al.,* 1999) by exposing the interior porous structures.

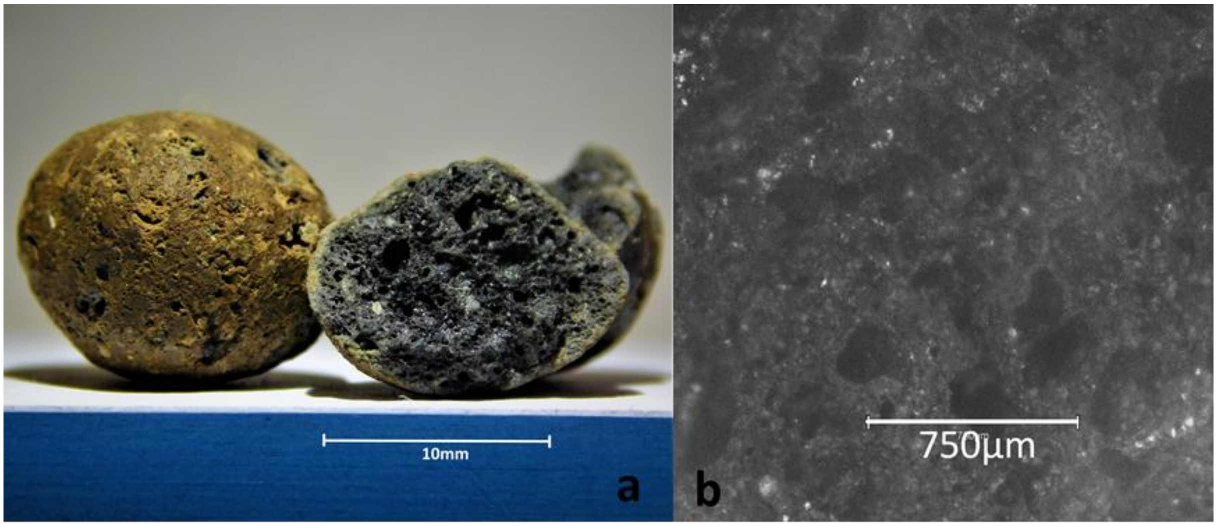

Figure 2 Macroscopic view (a) of intact and crushed LECA granule. Magnification of interior porous structures (b).

2.2 Performance of LECA in CWs

2.2.1 Removal of pollutants through adsorption

In LECA based CWs, especially in unplanted CWs, adsorption is one of the main routes for the removal of a wide range of water pollutants (Bahmanpour *et al.,* 2017; Białowiec *et al.,* 2009; Dordio and Carvalho 2013; Dordio *et al.,* 2007; Põldvere *et al.,* 2009).

Sharifnia *et al.,* (2016) did investigate ammonium adsorption to LECA and found the maximum monolayer adsorption capacity of LECA was 0.255 mg ammonium g^{-1}. The adsorption capacity was highest between pH 6-7, and equilibrium concentration was reached after 150 min with rapid adsorption within the first 60 minutes. High ammonium adsorption rates on LECA occur when the ammonium concentration in the water increases (Vymazal 2007).

Adsorption mechanisms of oxyanions such as phosphate occur via both anion exchange and ligand exchange (Yaghi and Hartikainen 2013; Yaghi and Hartikainen 2018). Phosphate is adsorbed as inner-sphere complex with the oxygen atom of phosphate bound directly to Al and Fe-oxides at the LECA surface (Kwon and Kubicki 2004; Zheng *et al.,* 2012). Inner-sphere complexes are considered strong and mostly irreversible (Yaghi 2015). Ligand exchange mechanisms occur preferably under acidic conditions, not only due to the positive surface charge at low pH, but also because of the increasing protonation of the OH^- groups at the mineral oxide surface, leading to the formation of aqua groups that swap more readily with oxyanions than OH^- groups (Yaghi 2015). To achieve high P removal it has been argued that LECA high in Al is preferable over high Fe, since Al sites are not redox-sensitive, retaining adsorption capacity at low redox potential, while Fe^{3+} might be reduced into Fe^{2+} which subsequently results in release of Fe bound P (Yaghi and Hartikainen 2013).

The Ca, Fe, Al, and Mg contents affect the amount of P adsorbed by LECA surfaces (Baker *et al.,* 2014). Among these elements, Ca has the strongest correlation with P-sorption capacity (Zhu *et al.,* 1997). A positive correlation was found between P removal and the content of both CaO and Ca in substrates (Vohla *et al.,* 2011). Therefore, low P removal in some LECA-based CWs (Table 2) can be attributed to the low Ca content of the substrate (Johansson 1997).

The pH is a critical parameter that affects the fate of phosphorous in CWs. The pH values in LECA beds can range from 4.0 to 9.5 (Mesquita *et al.,* 2013). Higher pH values have a positive effect on P adsorption and precipitation (Vymazal 2007). Previous studies indicated that an effective P removal in LECA based CWs occurs at high pH value ranging from 10 to 12 (Zhu *et al.,* 1997). The highest P adsorption (800 mg kg^{-1}) by LECA was achieved at a highly alkaline pH of 12.3 (Jenssen and Krogstad 2003; Zhu *et al.,* 1997), while only 72 hours were needed to reach the maximum adsorption capacity. The P adsorption in CWs involves two steps (Jenssen and Krogstad 2003). The first step is a short-term transition stage, mostly occurs at low P concentration and is barely affected by the CW operational regime. The second step leads to long-term binding and continues for weeks to months depending on substrate properties and P concentration. High P concentrations can depress pH and eventually the precipitation process of P. For optimal P adsorption by LECA, a retention time of 4 weeks is suggested (for colder climates) (Jenssen and Krogstad 2003).

LECA can have an influence on pH values of the water within the CW itself, because of its high contents of Ca minerals (Białowiec *et al.,* 2011). High pH values in the outflow of a LECA based hybrid CWs were measured with 8.1 to 8.8 in initial 9 months of operation and 7.6 to 7.7 in the three months following (Põldvere *et al.,* 2009). The highly alkaline conditions can adversely affect the growth of microbial communities which is important for organic matter and N removal processes (Tietz *et al.,* 2007).

LECA has a finite capacity to adsorb P. Its ceramic matrix makes LECA physically resistant but it is unlikely that new adsorption sites will emerge or generate in contrast to soil matrixes (Jenssen and Krogstad 2003). Beyond saturation surface accumulation of both organic matter and sediments may reduce LECA's adsorption capacity (Ballantine and Tanner 2010). LECA CWs can retain P through precipitation and sedimentation reactions with Ca-rich particles. The precipitation mechanism is favored at higher pH values or in presence of dissolved Ca in wastewater which promote P precipitation as Ca- phosphates especially during the initial stages of the treatment process (Jenssen and Krogstad 2003). However, when pH values and dissolved oxygen concentrations decrease further P precipitation is inhibited. Despite the significant contribution of wetlands sediments to P removal from wastewater, this P sink is often not considered in LECA based CW (Braskerud 2002; Mendes *et al.,* 2018).

Clays, in general, have good removal capacity for heavy metals due to their high cation exchange capacity (Ma and Eggleton 1999). This indicates that LECA has potential for heavy metals removal. LECA has been applied to remove Pb, Cu and Cd from industrial wastewater (Table 3) (Malakootian *et al.,* 2009) and mining tailings (Scholz and Xu 2002). Some anionic pharmaceuticals such as MCPA (4-chloro-2-methyphenoxyacetic acid), oxytetracycline and polyphenols can be removed by electrostatic interactions with LECA at neutral pH (Dordio and Carvalho 2013; Dordio *et al.,* 2007). In comparison, LECA showed better adsorptive removal for lipophilic compounds (oxybenzone and triclosan) compared to a hydrophilic compound (caffeine) (Ferreira *et al.,* 2017) and for polycyclic aromatic hydrocarbons (PAHs) including phenanthrene, fluoranthene and pyrenes (Nkansah *et al.,* 2012) (Table 3). The high removal rates in previous studies are attributed to LECA's exterior and interior surfaces that exhibit hydrophobic character, however, the underlying mechanisms are rather vaguely understood, as factors that provide hydrophobicity to LECA are not well addressed in the literature.

Table 3. Removal efficiency for heavy metals and organic contaminants using LECA substrates.

Contaminant	% Removal efficiency	Comments	Reference
Pb Cd	93.7 89.7	Short contact time ranging from 1 to 2 hours for Pb and Cd adsorption. The removal rate of Cd and Pb gradually decreased with increase in contact time. Adsorption occurred at pH ranging from 3 to 10.	Malakootian *et al.,* (2009)
Pb Cu	96 87	The presence of plants had no effect on Pb and Cu removal. Highest removal capacity observed for highly porous media.	Scholz and Xu (2002)
		Organic contaminants	
Oxytetracycline (antibiotic)	>97	Very high removal efficiency obtained in planted beds. Short contact time (within 3 days).	Dordio and Carvalho (2013)
Polyphenols	80	A large proportion was removed after 3 days of contact time in planted beds.	
MCPA (herbicide)	77	High removal obtained in planted beds.	
Caffeine (wastewater indicator) Oxybenzone (sunscreen agent) and Triclosan (anti-bacterial agent)	19-85 61-97	LECA showed high removal capacity for hydrophilic and lipophilic compounds.	Ferreira *et al.,* (2017)
Polyaromatic hydrocarbons (PAHs): Phenanthrene Fluoranthene Pyrene	 92 93 94	Suggested LECA as alternative method for PAHs removal.	Nkansah *et al.,* (2012)

2.2.2 Removal of pollutants through biological pathways

The main biological pathways for N removal in CW involve aerobic and anaerobic microbial metabolism through ammonification, nitrification and denitrification (Vymazal 2007) and both uptake and assimilation by plants and microorganisms (Wu *et al.,* 2011), while the substrate is a main parameters in determining both location and activities of the microbial community (Truu *et al.,* 2009). Previous studies have shown a decline in microbial density in the upper 10 cm of the substrate when porous materials such as sand and gravel were used as filtration bed (Braeckevelt *et al.,* 2007; Nurk *et al.,* 2005). The relocation of the microbial biomass into greater depths can be explained by the higher availability of organic matter and the shelter provided on the substrate surfaces and within the micropores between LECA grains (Calheiros *et al.,* 2009; Tietz *et al.,* 2007). LECA's capacity for high N removal has been attributed to its high porosity and large surface area (Saeed and Sun 2012; Vymazal and Kröpfelová 2009; Yang *et al.,* 2018), which allows oxygen to penetrate, especially if LECA is installed as an upper layer.

Plant roots in vegetated wetlands provide additional surface area for biofilm formation and growth, and create deep-reaching aerobic zones (Allen *et al.,* 2002; Brix 1993; Clairmont *et al.,* 2019; Gagnon *et al.,* 2007; Wu *et al.,* 2001). In vertical flow CWs, large proportion of the oxygen enters the substrate bed via diffusion, while in horizontal flow, the oxygen is mostly provided by the plants (Lee *et al.,* 2009; Molle *et al.,* 2006). Decaying roots provide readily accessible organic matter as additional carbon source and can remarkably improve denitrification rates (Lu *et al.,* 2009; Luo *et al.,* 2018). Planted LECA beds have been reported to have higher N removal capacity due to higher microbial diversity and density compared to unplanted ones (Almeida *et al.,* 2017; Białowiec *et al.,* 2009; Dordio and Carvalho 2013).

P uptake and assimilation by plants are the main biological routes for P removal in CWs (Kim and Geary 2001). The largest proportion of soluble P is taken up by microphytes and algae, especially in the early stages of the growing season. P uptake by plants contributes to a short term removal mostly during growth (Vymazal 2007) and if not removed decaying plants may lead to re-release of P into the wetland. Organic P which enters the CW as phospholipids, nucleic acids and sugar phosphates is transformed via the microbial metabolism. The microbial uptake of P is very fast and accounts for a temporary removal as microorganisms have a very short turnover rate (Qualls and Richardson 2000). Biological take-up of P in LECA based CW systems has not been quantified due to the dominance of P removal through adsorption.

The removal of organic matter i.e. BOD, chemical oxygen demand (COD) and total suspended solids (TSS) in CWs is driven by microbial degradation and the retention of these compounds to the substrate bed (Saeed and Sun 2012). LECA substrate has a good capacity for organic matter removal because of high porosity and specific surface areas which allow better biofilm adhesion to increase the biodegradation (Table 4). In a hybrid LECA CW, almost complete removal of BOD (99%) was achieved (Põldvere *et al.,* 2009; Zaytsev *et al.,* 2007). A high removal of COD (92%) and TSS (80%) was also reported by

Dordio and Carvalho (2013) in CW mesocosms with more than 60% of the organic matter removed by sedimentation on the LECA bed.

Table 4. The removal efficiency of LECA substrates integrated with different types of CWs for N, P and organic compounds from diverse types of wastewater

LECA/ other substrates	wastewater source	CW type	Planted/unplanted	Total N		NH4-N[1]		NO3-N[2]		Total P		TSS[3]		BOD[4]		COD[5]		HLR	HRT[6] (d)	Reference
				In	Out	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out			
				mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%	mg l^{-1}	%			
Bottom layer: 10 cm, LECA 10-20 mm. Middle layer: 25 cm, LECA 2-4 mm. Top layer: 10 cm LECA 3-8 mm	olive mill wastewater	vertical	planted	-	-	-	-	-	-	-	-	616	95	-	-	2160	92	-	6	Dordio and Carvalho (2013)
			Unplanted	-	-	-	-	-	-	-	-		95	-	-		81	-		
Bottom layer: 10 cm, LECA 10-20 mm. Middle layer: 25 cm, LECA 2-4 mm. Top layer: 10 cm	swine wastewater		planted	-	-	392	75.2	24	58.4	-	-	480	86	-	-	1420	80	-	9	
			unplanted	-	-		47.4		52.3	-	-		86	-	-		68	-		

LECA 3-8 mm																				
20 cm LECA 13-15 mm	synthetic wastewater	sequencing batch mode	planted	69	19	40	-35	-	-	19	18	-	-	-	-	203	55	-	48, 72 h	Lima *et al.,* (2018)
			unplanted		9		-32		-		25	-	-	-	-		47	-		
LECA	synthetic domestic wastewater	horizontal	planted	50	70	6	66	0.9	52	8	61	-	-	-	-	-	-	-	3	Özengin (2016)
			unplanted		65		57	0.9	66		67	-	-	-	-	-	-	-	-	
2-4, 4-10, 10- 20 mm	domestic sources and food processing plants	hybrid	unplanted	-	81	-	79	-	-	-	67	-	-	-	99	-	-	0.2-0.73 m^3 d^{-1}	-	Zaytsev *et al.,* (2007)
Limestone LECA		vertical filled with crushed limestone and a horizontal filled with LECA		-	82	-	83	-	-	-	60				99	-				
LECA 2-4, 4-10, 10- 20 mm; limestone	secondary treatment of domestic wastewater	hybrid	unplanted	72	47	-	-	-	-	20	66	132	94	405	82	745	64	52 mm d^{-1}	6	Põldvere *et al.,* (2009)
2–4 mm LECA	secondary treatment of domestic wastewater	batch mode	unplanted	54	82	-	-	-	-	6.6	48	33	82	135	99	224	70	59 mm d^{-1}	4	Põldvere *et al.,* (2009)
FASSTT with LECA and gravel	artificial wastewater	vertical	planted	-	59	-	99	-	-	-	-	-	-	-	-	-	-	4.6 mm d^{-1}	7	Białowiec *et al.,* (2011)
			unplanted	-	46	-	61–66	-	-	-	-	-	-	-	-	-	-			
4 to 8 mm	domestic wastewater	horizontal	unplanted	-	-	-	61-91	-	100	-	-	-	-	-	-	-	64-94	3.5 cm d^{-1}		Albuquerque *et al.,* (2009)
2-4 mm	pretreated domestic wastewater	horizontal	unplanted	-	-	-	-	-	83	-	-	-	-	-	60	-	-	-	1-4,7	Nurk *et al.,* (2009)
Filtralite® 4-8	synthetic	horizontal	planted	-	-	36.3	59.3	-	-	-	-	-	-	-	-	315.9	74	3.6 cm d^{-1}	6	Mesquita *et*

mm	wastewater		unplanted	-	-	26.7	33.9	-	-	-	-	-	-	-	-	311.2	38			*al.,* (2013)
LECA 10/20	synthetic wastewater	vertical	planted	-	-	-	83 mg l^{-1}	60	-	-	-	-	-	5300	-	82- 94 mg l^{-1}	-	148 to 473 L $m^{-2}d^{-1}$	-	Almeida *et al.,* (2017)
Filtralite® and gravel	tannery wastewater	horizontal	planted	-	-	-	-	-	-	-	-	-	-	*1800	*652	*3849	*1869	18, 8 and 6 cm d^{-1}	-	Calheiros *et al.,* (2008)
LECA 10/20 mm	domestic wastewater	hybrid constructed wetlands	unplanted	36.1	63	22.9	77			1.2	89	11.8	78	19	91			7.4 $m^3 d^{-1}$ to 17.7 $m^3 d^{-1}$		Öövel *et al.,* (2007)
LECA granules and powder	dairy industrial wastewater		unplanted						44.4		64.2	570	60	1220	68.4	2200	65.9		20 -120 h	Bahmanpour *et al.,* (2017)

*kg ha^{-1} d

The sedimentation of the organic matter occurs mostly near the CW inlet (Caselles-Osorio *et al.,* 2007). Organic matter accumulation is strongly correlated with organic loading rates (Meng *et al.,* 2015). The high average removal of both BOD (91%) and TSS (78%) in a vertical flow CW was attributed to the efficient mineralization of organic matter (Öövel *et al.,* 2007). The removal of BOD, COD and TSS was found to be affected by the vegetation type and the creation of aerobic zones within the rhizosphere which positively affected microbial density and metabolism (Lima *et al.,* 2018).

2.2.3 Pathogens removal

CWs have been increasingly adopted for wastewater reuse schemes, therefore pathogen removal has become a central treatment goal that determines wetland design and operation (Barbagallo *et al.,* 2010; Masi *et al.,* 2007).

CWs provide a number of biological, physical and chemical removal mechanisms for pathogens which mimic processes occurring in natural wetlands (Kadlec and Wallace 2008). driven by a combination of sedimentation and filtration, adsorption, predation, photoinactivation, natural die-off as well as biocidal effect of root exudates or internalization into plant tissue (Alufasi *et al.,* 2017; Boutilier *et al.,* 2009; Wand *et al.,* 2007; Wenk *et al.,* 2019; Wu *et al.,* 2016). Filter media in CWs contribute mostly to physiochemical pathogen removal mechanisms such as filtration and adsorption. Fine granular substrates trap microorganisms and increases their retention time by enhancing removal through natural-die off (Vacca *et al.,* 2005). Adsorption of pathogens was found to be particularly effective for substrates with positive surface charge (Rzhepishevska *et al.,* 2013). Both chemical composition and physical substrate properties, for example porosity, affect the microbial composition and biofilm growth and contribute to pathogen predation and adhesion (Long *et al.,* 2016; Meng *et al.,* 2014). However, the link between substrate properties, predation and microbial composition in CWs is currently not fully understood (Lee *et al.,* 2010; Mayes *et al.,* 2009). CWs for primary and secondary wastewater treatment operate at average influent *E.* coli concentrations of 10^5-10^8 colony forming units per 100 mL (cfu/100mL) for domestic wastewater (Headley *et al.,* 2013) and up to 10^{11} cfu/100mL for fecal coliforms in slaughterhouse wastewater (Rivera *et al.,* 1997). The typical removal rates of fecal microorganism observed in CWs range from 1-3 log units (Abou-Elela *et al.,* 2013; Headley *et al.,* 2013; Molleda *et al.,* 2008). In terms of water quality standards for water reuse, the free water surface systems located in tropical or subtropical climates are capable of producing final effluent with fecal-coliform concentration as low as 100 cfu/100 mL (Greenway 2005), while in temperate climates, the effluent could be consistently maintained around 1000 cfu/100 mL (Vivant *et al.,* 2016). Subsurface flow systems may achieve effluent concentration below 1000 cfu/100ml, particularly when employed as tertiary treatment step (Adrados *et al.,* 2018; Andreo-Martínez *et al.,* 2017). Nevertheless, many CWs exhibit high variability in effluent pathogen concentrations, and further research is needed to improve design towards a more consistent removal performance (Jasper *et al.,* 2013; Wenk *et al.,* 2019).

Due to the coarse granular size (5-20 mm), the water in LECA filtration beds has a relatively low residence time in comparison with sand beds, therefore bacterial adhesion mechanisms may not be very effective (Ausland *et al.,* 2002). Similarly, large granular size also excludes both filtration and straining from being an important removal mechanism in LECA-dominated systems (Díaz *et al.,* 2010). On the other hand, LECA's porous surface enhances biofilm growth and subsequent bio-clogging, which facilitates effective bacteria immobilization (Lianfang *et al.,* 2009). High cation exchange capacity of LECA could be also beneficial for bacterial removal since it enhances adhesion (Stevik *et al.,* 1999). Additionally, clay minerals in LECA, may alter i.e. metabolic pathways of biofilm microorganisms encapsulating the granule through increase of cell division in *E. coli* in the presence of kaolinite (Cuadros 2017). As a proven soilless plant growing substrate (Pradhan *et al.,* 2018), LECA may facilitate pathogen removal through root biofilm attachment (VanKempen-Fryling and Camper 2017) and possibly plant exudates (Alufasi *et al.,* 2017).

Consistent *E.* coli removal of 1.5 log-units was reported for a LECA-based horizontal flow polishing CWs after a prior filtration step, and the removal performance was similar to referenced gravel systems (Verlicchi *et al.,* 2009). Removal rates of up to 3 log for *E. coli* and total coliforms were reported in horizontal flow LECA CW (Calheiros *et al.,* 2015). Integration of LECA-based CW with preceding septic tanks may eliminate the dissemination of human parasitic helminth eggs (Paruch 2010). LECA upflow biofilters designed as unplanted subsurface CW, showed full removal of somatic coliphages which was attributed to the extensive attraction of negatively charged viruses onto the positively charged LECA surface (Heistad *et al.,* 2006). Due to the potential to reuse LECA as soil enhancer in agriculture, sanitation safety issues have been investigated. *E. coli* contamination of LECA from a horizontal flow CW persisted for more than 14 months after the last contact with wastewater (Paruch 2011). However, despite the long survival time, *E. coli* concentrations below 2.5 10^3 cfu/g of dried substrate, allowed reuse for agricultural applications based on regulatory requirements (Paruch *et al.,* 2007). Survival of coliform bacteria on LECA has been tested to assess the health hazards related to the use of vertical flow CW in densely populated areas (Bydałek and Myszograj 2019). When exposed to atmospheric conditions as a top filtration layer in vertical flow CWs, LECA showed slower inactivation rates of coliforms (k_{6h}=0.36h^{-1}, k_{12h}=0.25h^{-1}) in comparison to gravel or slag but faster inactivation compared to organic substrates such as bark and charcoal.

2.2.4 Modified LECA materials

Sorption in CWs is a finite process, that requires periodic exchange of the wetland substrate (Arias and Brix 2005; Drizo *et al.,* 2002). Efforts to extend CW sorption performance have been focusing on substrates with improved P removal, CW management including hydraulic operation practices and both ex-situ and in-situ substrate treatment (De la Varga *et al.,* 2013; Knowles *et al.,* 2011; Lianfang *et al.,* 2009; Nivala and Rousseau 2009; Pedescoll *et al.,* 2009).

LECA can be improved through changing its mineral composition or altering its surface charges via coating or additives (Table 2). Coating LECA with Al, Fe or Mg oxides, has indicated a positive effect on P, As and pharmaceutical removal, respectively (Haque *et al.,* 2008; Kalhori *et al.,* 2017; Yaghi and Hartikainen 2013; Yaghi and Hartikainen 2018). Lime had a positive effect on P adsorption capacity (Johansson 1997). Mixing raw materials with fly ash and dolomite was found to enhance P and N removal capacity, hydraulic conductivity, and porosity (Białowiec *et al.,* 2011; Jenssen and Krogstad 2003). Adding of sodium carbonate (Na_2CO_3), quartz (SiO_2), hematite (Fe_2O_3) or elemental iron (Fe) at 2-10 wt% into the raw clay increased LECA density, porosity and crushing strength (Bernhardt *et al.,* 2014), while added quartz sand altered particle size distribution and the internal structure of the LECA by refining gas release during the expansion process (Fakhfakh *et al.,* 2007).

Bioaugmentation has been investigated to enhance denitrification and pollutants removal in LECA based CWs. The studies argued that LECA is a sterile substrate given its high temperature manufacturing process, while the microbes received by the influent provide insufficient capability to evolve an efficient treatment process. Introducing an already adapted microbial culture to a newly established CWs could positively affect performance (Nurk *et al.,* 2009; Zaytsev *et al.,* 2011), leading to a faster achievement of treatment goals. Augmentation of white-rot fungus *Lentinula edodes* to inoculate LECA and other substrates including cork and straw and coat pine enhanced pesticide degradation by almost 50% (Pinto *et al.,* 2016). Bioaugmentation has been assessed for many years for wastewater treatment applications. However, the impact on treatment performance is rather difficult to predict compared to the earlier mentioned chemical and physical modification strategies (Herrero and Stuckey 2015).

Table 2. Coatings and additives used for LECA properties and adsorption capacity improvement.

Coatings/ Additives	Treatment	Effects	Reference
Fe and Al oxides	As in groundwater	High adsorption capacity for As ions obtained at pH 2, 4 and 6.	Yaghi and Hartikainen (2018)
Fe oxide	As in groundwater	Faster adsorption, including increased capacity. Maximum As accumulation 3.31 mg of g^{-1} LECA at pH 6 to 7.	Haque *et al.,* (2008)
Fe and Al oxides	P in groundwater	High adsorption capacity. Al-coated sorbents were superior to Fe-coated ones.	Yaghi and Hartikainen (2013)
MgO nanoparticles	Pharmaceuticals: metronidazole antibiotic	High specific surface area (76.12 m^2/g). Antibiotic adsorption increased by approximately 33% as adsorption sites increased.	Kalhori *et al.,* (2017)
TiO_2 photocatalyst	Ammonia	High removal efficiency. The maximum degradation of NH_3 occurred at pH 11.	Zendehzaban *et al.,* (2013); Shavisi *et al.,* (2014)
TiO_2 sol-gel photocatalyst	Pharmaceuticals: tetracycline antibiotics and doxycycline	Improved mechanical stability Satisfactory photocatalytic antibiotic oxidation efficiency.	Pronina *et al.,* (2015)

Fe/TiO_2 and Cu/TiO_2 photocatalysts	Phenol from synthetic wastewater	61 % degradation of phenol in synthetic wastewater.	Sohrabi and Akhlaghian (2016)
TiO_2 /Zinc oxide (ZnO)/LECA hybrid photocatalyst	Ammonia from synthetic wastewater	95.2% of ammonia removal during the first 3 hours.	Mohammadi *et al.,* (2016)
H_2O_2-modified LECA	Water contaminated with fluoride	Increased surface area and adsorption capacity.	Sepehr *et al.,* (2014)
$MgCl_2$-modified LECA		Fluoride adsorption capacities of 17.83 mg/g and 23.86 mg/g for H_2O_2-modified LECA and $MgCl_2$-modified LECA respectively.	
Na_2CO_3 SiO_2 Fe_2O_3	Physical characteristics of LECA	Decreased viscosity of the surface. No effect. Pore size increased and density reduced.	
LECA made of fly ash from sewage sludge		Promoted activity of a consortium of micro-organisms responsible for N removal. Provided loose, porous, and well-aerated substrate thus nitrifying bacteria prefer to attach to it. Increased N removal efficiency.	Białowiec *et al.,* (2011)
$CaCO_3$/ Lime	P removal capacity	P adsorption increased	Johansson (1997)
Dolomite	N, P removal capacity and physical characteristics	High N and P removal. Enhanced LECA hydraulic conductivity, porosity and its insulation properties.	Jenssen and Krogstad (2003)

Quartz sand (< 250 µm grain size) and 1% motor oil (expansion promotor)		Better expansion properties. Physical properties such as apparent density and mechanical resistance improved.	Fakhfakh *et al.,* (2007)
		Biological additives	
Bioaugmentation in a newly established LECA-based horizontal flow	Biochemical process	Change in the structure of the microbial community. High performance and stable denitrification process.	Nurk *et al.,* (2009); Zaytsev *et al.,* (2011)
Bioaugmentation using white-rot fungus *Lentinula edodes*	Pesticides group: terbuthylazine, difenoconazole, diflufenican and pendimethalin.	Moderate retention capacity of pesticides. Microbial activity enhanced by porosity.	Pinto *et al.,* (2016)

3. Design considerations for LECA-based CWs

3.1 Layout of CWs using LECA substrate

In the majority of CW designs the substrate is arranged into horizontal layers (Kadlec and Wallace 2008). In larger more heterogeneous treatment wetlands with various sections or consecutive treatment cells different types of substrate may be used spatially (Lu *et al.,* 2016). Simple design CWs contain a single substrate, that is usually confined by an impermeable bottom liner (Almeida *et al.,* 2017) such a design is particularly common in decentralized, rural areas, where CWs serve single households (Figure 3).

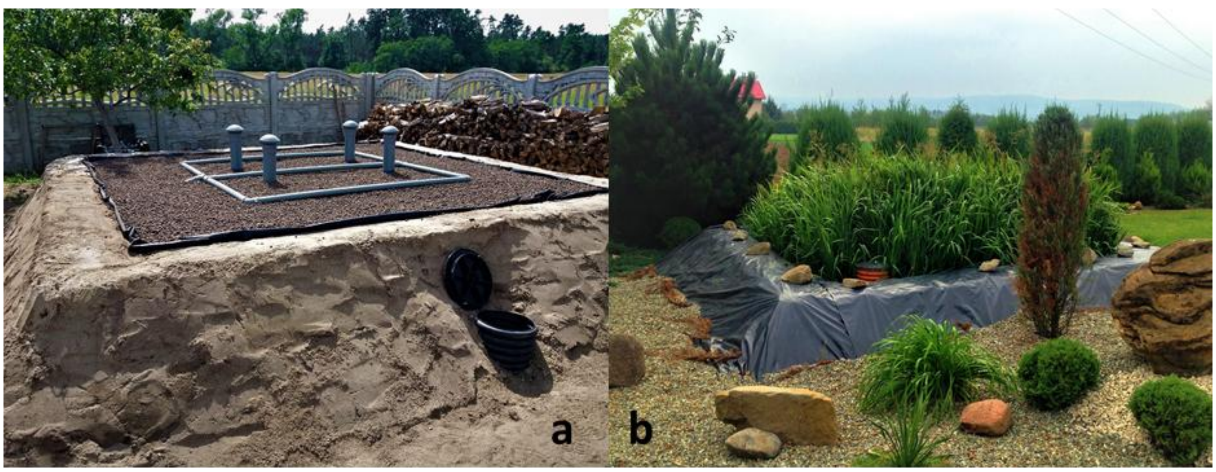

Figure 3 LECA-based Vertical Flow Constructed Wetland before (a) and after (b) commissioning. LECA has become one of the most commonly applied substrates for small scale, domestic CWs systems, which account for roughly 3000 units in Poland alone (*personal communication*). (Photo. F. Bydalek/ Ecoverde Engineering Office, Poland).

Multi-layered wetlands have been constructed with up to three different layers, while double layers are most common (Vymazal 2013b). Using double layers in vertical flow may create different oxic conditions as nitrifying bacteria prefer to attach to porous and well aerated media, whereas denitrifying bacteria colonize more compact aggregates that support low oxygen conditions (Białowiec *et al.,* 2011). Multilayers can be exclusively composed of LECA granules of different grain sizes or incorporate different types of substrates (Białowiec *et al.,* 2011; Calheiros *et al.,* 2009). Horizontal positioning of different substrate layers is variable. LECA has been mostly used as the upper layer when applied with other substrates to remove suspended solids and promote the growth of nitrifying microorganisms while providing aeration (Almeida *et al.,* 2017). On the other hand, installing LECA as a bottom layer substrate has a positive effect on the hydraulic conductivity and protects the system against clogging (Suliman *et al.,* 2006). Layer arrangements uniformity and grain size distribution within each layer are also critical for adequate hydraulic conditions to minimize clogging issues (Brix *et al.,* 2001). The grain sizes used in LECA beds can range from smaller 1 mm (powdery form) to 10/20 mm, sizes of 2/4, 3/8, 4/10 and 13/15 mm have also been applied for different types of CWs (See table 4). Different depths of LECA layers were tested to compare performance with thicknesses ranging from 12 cm to 150 cm in lab trials using columns or mesocosms with narrow volumes e.g. 0.25 m^2 (Almeida *et al.,* 2017; Białowiec *et al.,* 2011; Nurk *et al.,* 2009; Özengin 2016). LECA layer depths ranging from 20-90 cm have been used in a three layer hybrid CW of an area of 216 m^2 for domestic wastewater treatment (Öövel *et al.,* 2007). For the vertical flow section of this wetland a layer of 50 cm of coarser granules 10-20 mm was used as bottom layer covered by 30 cm of finer 2-4 mm granule to ensure oxygen transport. The vertical bed was followed by a horizontal subsurface flow filter (90 cm in depth), filled with 2-4 mm LECA granules. Installing multiple layers of LECA with coarser granules ranging from 10-20 mm can maintain a good hydraulic conductivity (Põldvere *et al.,* 2009).

Constructed wetlands have been used for the treatment of a wide range of different types of water including domestic, agricultural and industrial sources (Vymazal 2009). For example, dairy farm and aquaculture effluent can be high in COD, proteins, N species and phosphate (Dauda *et al.,* 2019; Justino *et al.,* 2016; Nagarajan *et al.,* 2019), and greenhouse effluent is usually high in nitrate (Prystay and Lo 2001). The composition of domestic wastewater is usually more similar across different locations (Tran *et al.,* 2015). Typical values of main wastewater parameters to size CWs were proposed by Kadlec and Wallace (2008): BOD 220 mg l^{-1}; TSS 500 mg l^{-1}; TN 40 mg l^{-1}; and P 8 mg l^{-1}.

Physicochemical properties of LECA (Figure 4) make it suitable for application in domestic wastewater treatment, targeting bioavailable N species, organic matter and P (Albuquerque *et al.,* 2009; Lu *et al.,* 2016; Meng *et al.,* 2015; Özengin 2016). For this type of wastewater LECA containing CWs have achieved a maximum reduction up to 99% BOD, 94% TSS, 83-99% ammonium and 89% P (see Table 4). Organic matter removal in LECA based CWs is significant for all types of wastewater. LECA has shown a relatively good capacity for P removal from domestic and food processing wastewaters with values ranging from 60% to 67.3% (Özengin 2016; Põldvere *et al.,* 2009; Zaytsev *et al.,* 2007).

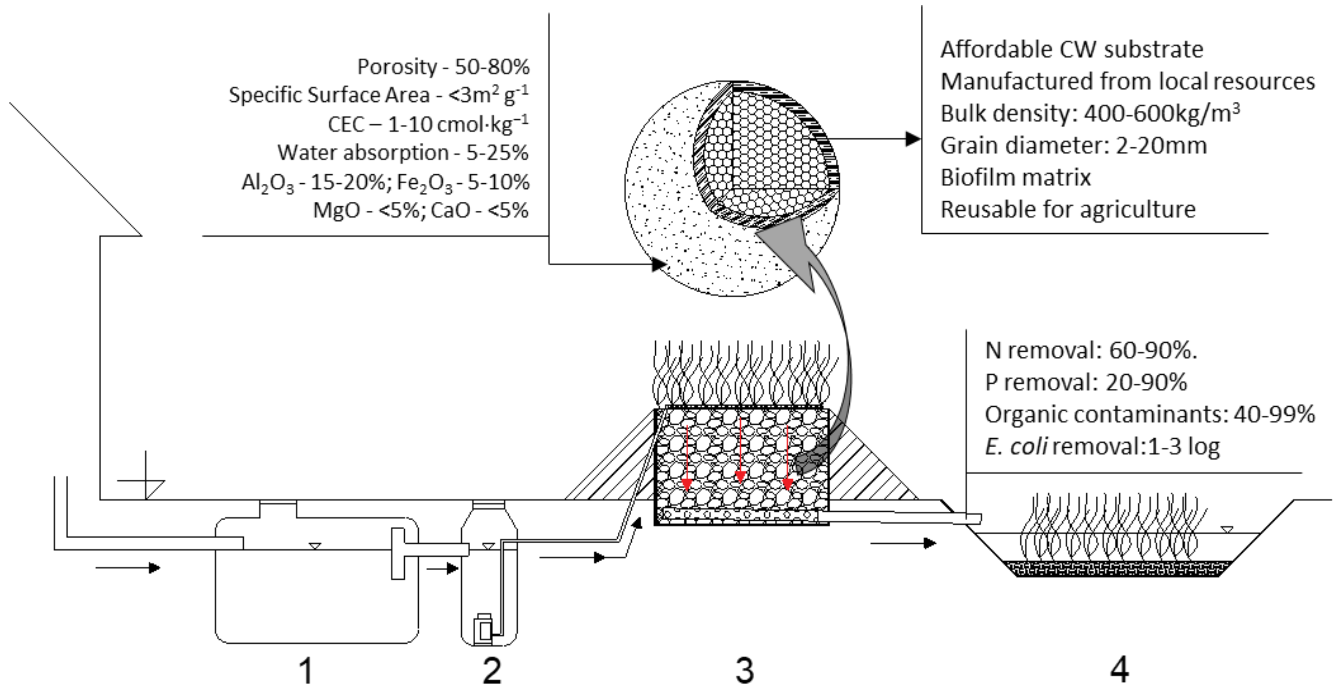

Figure 4 LECA properties enhance biological and physiochemical pollutant removal pathways in CWs. Schematic presentation of LECA-based VFCW designed for household use- 1) septic tank, 2) pump well, 3) elevated VFCW and 4) polishing pond.

LECA substrate has been also applied to remove heavy metals from urban runoff and a wide range of industrial wastewater including mining tailings, tanneries and dye factories (Calheiros *et al.,* 2008; Malakootian *et al.,* 2009; Scholz and Xu 2002) and agricultural wastewater including olive mill effluent and swine wastewater (Dordio and Carvalho 2013). Accumulation of organic matter and clogging at the inlet of CWs is a major challenge for high COD treatment tasks (Healy *et al.,* 2007; Langergraber *et al.,* 2003). LECA substrates ranging from 8-10 mm size have been shown to facilitate clogging issues, while smaller sized LECA substrates of 1-4 mm could not prevent clogging efficiently (Albuquerque *et al.,* 2009; Suliman *et al.,* 2006). Pre-dilution of raw wastewater before being introduced to the CW coupled with using fine particles

(2-4 mm) can minimize the clogging problem resulting from the accumulation of organic matter (Dordio and Carvalho 2013).

3.2 Hydraulic loading rate and hydraulic retention time

The hydraulic conditions such as loading rate and retention time are vital factors determining the treatment process in CWs (Ghosh and Gopal 2010; Jing *et al.,* 2002; Persson *et al.,* 1999). The hydraulic loading rate should be balanced with the expected oxygen depletion along the wetland (Liu *et al.,* 2016). Generally, low hydraulic loading rates and increasing hydraulic retention times lead to greater nutrient removal efficiency (Almeida *et al.,* 2017), whereas organic overloading results in hydraulic dysfunctions via clogging (Knowles *et al.,* 2011). Herrmann *et al.,* (2013) found that a loading rate of 100 L m^{-2} d^{-1} increased the average P binding capacity of LECA wastewater filters to 1.1 g kg^{-1} at residence times ranging from 5 to 15 min. High removal capacity of P in LECA beds is attributed to the hydraulic conductivity and the adaptability of LECA to changing hydraulic loads (Öövel *et al.,* 2007). Effluent recirculation enhances nitrification processes through increasing both the contact time of wastewater with CW biofilms and the supply of oxygen and organic matter into the wetland (Saeed and Sun 2012). Effluent recirculation has been tested for a hybrid LECA CWs, it was found that high recirculation rates of up to 300% in a hybrid CW can increase removal efficiency for BOD, TSS, total N (Table 4) (Põldvere *et al.,* 2009; Zaytsev *et al.,* 2007). A hydraulic loading rate of 239 ± 7 L $m^{-2}d^{-1}$ at a hydraulic retention time of 140 min was found to increase nitrate removal by maximum 66%, any further increase in hydraulic loading rate was found to have an opposite result on nitrate removal rate (Almeida *et al.,* 2017). Dordio and Carvalho (2013) indicated that

LECA adsorption capacity in planted beds was most effective after 6 days for TSS (95.3%), and COD (92.5%) and 9 days for ammonium (75.2%) and nitrate (58.4%).

3.3 Dissolved oxygen

The oxygen concentration of influent wastewater can range from almost anoxic (0.6 mg l^{-1}) to almost saturated (7.8 mg l^{-1}) levels (Liu *et al.,* 2016). Complete oxygen depletion in CWs is nevertheless common when treating high organic or N loaded wastewaters (Albuquerque *et al.,* 2009). The depth of the filtration bed influences DO distribution within CWs. In vertical flow CWs more than 90% of the oxygen penetrates the system by air diffusion; most of it is consumed by BOD removal and nitrification processes in the upper zone (Li *et al.,* 2014). Porous, large grained and loose substrates enhance oxygen transfer into the filtration bed (Verhoeven and Meuleman 1999). Nevertheless, despite of LECA porosity, low DO concentrations have been an issue similar to other types of substrates (Mesquita *et al.,* 2013). The reported values are ranging from 0.5 mg O_2 l^{-1} to 1.5 mg O_2 l^{-1} (Albuquerque *et al.,* 2009; Lima *et al.,* 2018) which is the minimum DO concentration required for nitrification. Many studies indicated that shorter hydraulic retention time ranging from hours to a few days can create favorable conditions for efficient use of oxygen by the microbial biomass. High DO fluxes may result in weak denitrification (Shuib *et al.,* 2011; Tao *et al.,* 2006; Xiao *et al.,* 2010). Oxygen transfer into CWs, can be enhanced via vegetation (Li *et al.,* 2014; Vymazal and Kröpfelová 2008) and management of hydraulic conditions in addition to active aeration (Liu *et al.,* 2016; Ouellet-Plamondon *et al.,* 2006). In LECA based CWs effluent recirculation can improve aeration conditions and overall purification efficiency (Põldvere *et al.,* 2009) such as BOD removal

(Zaytsev *et al.,* 2007). Alternatively, batch (drain and fill) feed mode can create more oxygen-rich conditions compared to continuous feed mode, and increase N, P and COD removal (Zhang *et al.,* 2012).

3.4 LECA CWs under different climatic conditions

CWs have been operated under a variety of climate conditions (Jenssen *et al.,* 2005; Koottatep *et al.,* 2005; Quanrud *et al.,* 2004). Cold climate can significantly affect hydraulic performance and both biological and chemical processes in CWs; microbial activity and vegetation growth are reduced at low temperatures (Werker *et al.,* 2002). The N removal is reported to be inhibited below 10 °C (Luo *et al.,* 2005) and nitrification does not occur below 4°C (Cookson *et al.,* 2002). A decrease in water temperatures from 20 to 5°C was found to decrease the adsorption capacity of LECA by 24% to 64%, increasing with grain size (Zhu *et al.,* 1997). Design alternations to improve wetland performance in cold climates include lower hydraulic loading and both selection of tolerant vegetation and specific substrates (Yan and Xu 2014). LECA has been extensively used for CWs in cold climate (Brix *et al.,* 2001; Jenssen *et al.,* 2005; Johansson 1997; Mæhlum 1995; Suliman *et al.,* 2006), however in subtropical and semiarid climates i.e MENA (Middle East and North Africa) region, use of LECA as a substrate in CWs is rather absent or not addressed in the literature.

4 Recycling of wetland substrates and environmental concerns

CWs have become an accepted and established technology for the treatment of water. The fate of the wetlands substrates after saturation is rather vague and poorly addressed in the literature (Jenssen and Krogstad 2003; Johansson Westholm 2006). More recently concerns have been raised about the fate of the substrates after the end of their useful lifetime (Yang *et al.,* 2018). Substrates upon saturation may contain high concentrations of nutrients, organic compounds and in some cases, toxic contaminants and pathogens (Hench *et al.,* 2003). Many studies highlighted the possibility of using spent LECA from CWs as P fertilizer and soil liming amendment for acidic soils (Jenssen *et al.,* 2010; Johansson Westholm 2006; Vohla *et al.,* 2011) considering LECA's P adsorption potential which can reach up to 12,000 mg P kg^{-1} (Ádám *et al.,* 2006; Ádám *et al.,* 2007). However, P saturated LECA may not support short term P release in soils, including availability to plants. Hylander and Simán (2001) did test different types of saturated substrates and found that P-saturated LECA resulted in lower crop (barley) yields compared to crystalline slag substrates. In LECA P was bound tightly to Al and Fe oxides, while the P in slag was bound to Ca and more readily available to plants.

Production of LECA is known to have a high energy demand (Johansson Westholm 2006), but actual quantitative information is virtually absent in literature, we only found one website based reference. This data indicated that amount of energy needed for producing 1 m^3 of LECA was estimated to be 931 MJ, while the CO_2 emission potential was 54 kg for the same quantity (www.leca.com). Therefore LECA is considered a high energy consumption manufactured substrates, its costs are determined by the production process rather than by the raw materials (Ballantine and Tanner 2010). Sustainable solutions for recycling and regeneration of LECA are needed to manage its fate and minimize energy consumption.

5 Future research directions

LECA is an adsorptive material that has a high removal capacity for Phosphorus (P) compared to other types of constructed wetland substrates. Beyond P, interactions of LECA with wastewater contaminants including organic trace contaminants, certain pathogens, in particular viruses, but also the nitrogen (N) species ammonium and nitrate need further investigation. Although, N removal in CWs occurs mainly through biological routes, substrates such as LECA may provide a buffer capacity, when metabolic processes temporary slowdown. Modification to tailor LECA for specific use in CW applications for better performance of desired treatment tasks or to improve biofilm development, including addressing clogging issues has untapped potential. Such modified properties might be achieved through relatively simple means by crushing pellets to expose the inner structures or by blending additives into raw clay mixtures. There is need to develop reuse and recycling strategies for spent CW substrates, including opportunities for P recovery, while considering potential heavy metals and pathogen loads. The energy required during LECA production needs to be accounted for when assessing its life cycle in comparison with alternative substrates.

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Response letter to the editor

Date: 20/01/2020

Editor-in-Chief: Dr. Jan Vymazal

Journal name: Ecological Engineering

Dear Dr. Jan Vymazal

Subject: submission of the revised review manuscript "Light-expanded clay aggregate (LECA) as a substrate in constructed wetlands–A review"; manuscript ID number ECOLENG-D-19-00882R1.

Thank you for your email dated 20/11/ 2019 enclosing the reviewers' comments. We would like to thank the two reviewers for their time and their valuable and positive comments that helped us to improve the manuscript. We have carefully reviewed the comments and revised the manuscript accordingly. Before addressing the reviewer comments specifically we would like to draw your attention to the main changes that we applied to the manuscript.

- We have addressed the major point of the reviewers to minimize the length of the review, and reduced the text by almost 1/3. We found that further reduction to ½ as suggested would have weakened the review by removing too much of the context and resulting in decreased readability.
- We consolidated the text to make it appear less fragmented and have integrated the previous subsections: ammonium adsorption, phosphates adsorption, and heavy metals and organic pollutants were removed and into one subtitle: Removal through adsorption.
- The subsections biological nitrogen removal; biological removal of P and organic matter removal were integrated into one subsection: Removal through biological pathways.
- The pathogen removal section was shortened and moved towards a later section of the text.
- The modified LECA materials subtitle (2.2.4) was moved to the end of the performance of LECA in CWs (2.2) section as we saw best to present the performance of the classical LECA in CWs and then the modified one later.
- Authors list has been updated in line with the final contribution to the revised version.
- Tables and figures have been integrated into the manuscript body next to the relevant text.

Responses to specific Reviewer comments. The reviewer comments are presented in *Italic*, followed by our response.

Reviewer #1: '*The overall quality of this review is fairly good but it does appear to be a length work.'*

- We would like to thank reviewer 1 for their positive evaluation of the manuscript.

'The content should be fully focused on the use of LECA as a substrate in constructed wetlands. Background information, overly described in the current version, should be given in a pithy style. For example, the basic information about constructed wetland, the original use of LECA and its production, the basics of biological nitrogen removal pathway and the role of vegetation.'

- Basic information in both introduction and in the main body, including nitrogen removal pathways and the role of vegetation of the text has been either removed or considerably shortened.

'One obvious mistake is the wrongly numbered tables. Table 4, appearing first, should be replaced to Table 1. The order of Table 3 and Table 2 should be exchanged.'

- We apologize for the errors, these are now corrected. The tables' numbers have been switched so they appear in order in the text.

'Compared with Table 2, more details (e.g., CW type, operating conditions, influent concentration of heavy metals/organic contaminants) should be added in Table 3 making it more informative.'

- We have tried to integrate those tables, but similar information to table 3 components was not available in the literature cited, thus the table would appear fragmented if integrated.

Regarding the design considerations for LECA-based CWs, the content should be more specifically related to the special properties of LECA.

- We believe that the content of LECA-based CWs was already properly addressed in the review, we therefore did not make changes, also regarding the need to reduce the overall length of the manuscript. Two examples on the effect of LECA granules size on some operational parameters in LECA based CWs are provided below:

- ✓ *Line 454-456 "Layer arrangement uniformity and grain size distribution within each layer are also critical for adequate hydraulic conditions to minimize clogging issue".*
- ✓ *Line 463-465 "For vertical flow section of this wetland a layer of 50 cm of coarser granules 10-20 mm was used as bottom layer covered by 30 cm of finer 2-4 mm granule to ensure oxygen transport."*

'Since the conveyed information from Figure 1 and 4 were overlapped to some extent, these two figures are suggested to be combined to one. The same suggestion was made for Figure 2 and 5.'

- Figure 1 has been deleted and the content has been merged. We tried to combine Figures 2 and 5 (now 1 and 4), however, the resulting merged figure appeared rather complex and therefore, we decided to keep the figures separately. Figure 2 shows more general CW concepts, while Figure 4 provides more LECA specific content.

'Besides, the performances of LECA-based CWs should be critically compared with those CWs using other special substrates.'

- The current review focused more on the specific issues and considerations of LECA based system, we thus limited the extent of the comparison with other special substrates. Note that other reviews cited in our review include comprehensive comparisons between different types of natural, man-made and by-products substrates. However, in these reviews more general information about LECA have been presented, LECA thus was researched somewhat superficially and less in-depth compared to our review:

See also introduction (125-140): Recent reviews have compared the role of different substrates on the removal of nutrients, including P removal of both natural and manufactured substrates (Wang et al., 2020) and summarizing the structural differences and inherent properties of unconventional substrates such as zeolite, rice husk, alum sludge among many others, and their capacity for substantial N and organics removal from wastewater under optimized operating conditions (Saeed and Sun, 2012). Various substrates have been classified based on their ion-exchanging, P sorbing and electron donating properties (Yang et al., 2018). In general, substrates rich in mineral oxides of calcium (Ca), aluminum (Al), and iron (Fe) such as limestone, biotite, muscovite, steel slag, and light weight expanded clays aggregates (LECA) have high capacities of P and N removal (Johansson Westholm, 2006), while organic substrates such as rice straw, compost, and wood mulches can be utilized by microbes as electron donors and thus enhance nitrification and denitrification processes (Cao et al., 2016). Specific studies have focused on substrates with extensive capacity for P removal such as clay bricks, fly ash, wollastonite, slag material, bauxite, shale, burnt oil shale, limestone, zeolite and LECA (Drizo et al., 1999; Johansson Westholm, 2006; Lima et al., 2018).

Therefore, we believe the Reviewer comment should be sufficiently well-addressed, while the interested reader is referred to other reviews.

Reviewer #2: *'Thank you for the effort in putting together all this very complete description about the usage of Leca in Treatment Wetlands. Very well conducted review.'*

- We would like to thank the Reviewer for the positive evaluation of our manuscript.

'If the Editors will consider the length of the paper excessive, I am sure that the paper can be shortened of 1/3 or even more than 1/2 providing less details for the several applications, especially not repeating same explanations of similar findings many times.'

- We agree with the reviewer that the initial version of the manuscript was lengthy in parts. Therefore, the manuscript has been shortened by ca. 29%. Further reduction was felt to decrease the quality of the review.

We have now uploaded the new version of the manuscript and an annotated original version of the manuscript that contains major deleted section in yellow. Note that due to the major changes conducted by all authors collaboratively, including shifting references, it was not possible to upload a tracked changed version of the initial manuscript. In addition we have conducted minor shortenings throughout the text, removed typos and improved both grammar and language.

We hope we have addressed the reviewers' comments adequately well and look forward to hearing from you in due course.

On behalf of all authors
Kind regards

Rawan Mlih
PhD student
Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Germany.
Institute for Environmental Research, Biology 5, RWTH Aachen University, Germany

Light-expanded clay aggregate (LECA) as a substrate in constructed wetlands–A review

Rawan Mlih[*1, 6], Franciszek Bydalek[2, 3], Roland Bol[1], Nader Yaghi[4], ~~Rob van Deun[5],~~ Erwin Klumpp[1, 6], Jannis Wenk[3]

[1] Institute of Bio- and Geosciences, Agrosphere (IBG–3), Research Centre Juelich, Wilhelm Johnen Str., 52425 Juelich, Germany

[2] GW4 NERC Centre for Doctoral Training in Freshwater Biosciences and Sustainability, Museum Avenue, Cardiff CF10 3AX, United Kingdom

[3] Department of Chemical Engineering, Water Innovation & Research Centre (WIRC), University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom

[4] Department of Chemistry, Faculty of Sciences, University of Hebron, 90100 Hebron, Palestine

~~[5] Thomas More University of Applied Sciences, Kleinhoefstraat 4, 2440 Geel, Belgium~~

[6] Institute for Environmental Research, Biology 5, RWTH Aachen University, Worringerweg 1, 52074 Aachen, Germany

*Corresponding author: r.mlih@fz-juelich.de

Abstract

Light expanded clay aggregates (LECA) have been increasingly used as substrate material for constructed wetlands given their phosphate removal capacity, mechanical strength, hydraulic conductivity and their plant rooting and biofilm growth supporting structure. This review summarizes the current literature on LECA-based constructed wetlands. Removal performances for main wastewater parameters phosphate, nitrogen species, suspended solids and oxygen demand are tabulated. Both, physical and biological water purification processes in LECA wetlands are discussed. Additional emphasis is on design and layout of LECA wetlands for different types of wastewater, under different climatic conditions and to improve treatment performance in general. LECA life cycle considerations include sourcing, production energy demand, reuse and recycling options for spent wetland substrates, for example as soil amendment. Research and development opportunities were identified for structural and compositional LECA modification to obtain tailored substrates for the use in water treatment and specific treatment tasks. Beyond traditional wastewater contaminants the fate of a wider range of contaminants, including organic trace contaminants, needs to be investigated as high Fe, Al and Ca oxides content of LECA substrates provide adsorptive sites that may facilitate further biological interactions of compounds that are otherwise hard to degrade.

Keywords: Constructed wetlands, LECA, pollutants removal, phosphorous, nitrogen, adsorption

Introduction

Preventing water scarcity, increasing water security and addressing water pollution are key actions to implement United Nations sustainable development goals (UN, 2015). The deterioration of water quality, including the decline of natural water resources, due to agricultural, industrial and domestic human activities is a global issue (Famiglietti, 2014; Vörösmarty et al., 2010). To maintain water quality and to protect aquatic habitats polluted water needs treatment before being released into natural water bodies or being reused. Traditional water treatment strategies employ a combination of physical, chemical and biological methods and require large investments in both infrastructure and operation (Goel, 2006; Hendricks, 2016; Sedlak, 2014). Managed natural systems such as constructed wetlands (CWs) are considered as a viable alternative to conventional treatment systems. CWs can serve for a wide range of treatment targets and contaminants at comparable removal efficiency to conventional treatment, while being less expensive to build, including lower energy demand and maintenance costs (Vymazal, 2010) (Figure *1*). CWs are artificial wetlands for wastewater treatment, they consist of a flow-through substructure, saturated with water and are planted with adaptive vegetation (Verhoeven and Meuleman, 1999).

Similar to natural wetlands, CWs have been recognized for their multiple roles that combine environmental and societal benefits, including improving water quality, increasing water storage buffer capacity during draughts and storm events, restoring wildlife habitats and providing diverse recreational space within the urban landscape

(Thorslund et al., 2017). Since the introduction of the concept more than 50 years ago (Seidel, 1961), the technology has advanced and CWs have been successfully used to treat an extensive range of domestic, agricultural and industrial wastewater streams under various climatic conditions (Calheiros et al., 2007; Merlin et al., 2002; Rozema et al., 2016). CWs have been integral part of progressive ecological urban planning for example in the 'sponge city concept' and are well-established in decentralised water treatment schemes for smaller communities and rural settlements (Arheimer et al., 2004; Liu et al., 2017).

There are three major types of CWs (Wu et al., 2015a): free surface water flow CWs, horizontal subsurface flow CWs and vertical subsurface flow CWs (Figure *2*). Free surface water flow CWs closely replicate the natural cleaning processes occurring in natural wetlands and have been applied for different types of wastewater including those with high biological oxygen demand (BOD) and solids content (Ghermandi et al., 2007; Vymazal, 2013a).

~~Both horizontal and vertical subsurface CWs are widely used (Luederitz et al., 2001), while hybrid systems may combine advantages of each type of CW (Vymazal, 2010). CW design is determined by specific treatment tasks, for example, free water surface CWs, which typically consist of a shallow basin, are appropriate for water with high solids content such as mining drainage, storm water and agriculture runoff (Dal Ferro et al., 2018; Niu et al., 2016). Subsurface flow CWs are suitable for water with low solids contents due to the hydraulic constraints imposed by the substrate (Vymazal and Kröpfelová, 2009).~~ CWs range from simple, vegetated soil filtration beds to highly diverse multi-hectare systems that combine different types of CWs (Dunne et al., 2012; Wu et al., 2015a). The removal mechanism of pollutants in CWs is achieved through an integrated combination of biological, physical and chemical interactions among plants, the wetland substrates and microorganisms (Truu et al., 2009; Vymazal, 2005). For instance, nitrogen (N) removal is achieved by microbial processes such as ammonification, nitrification, and denitrification (Vymazal and Kröpfelová, 2009), while physicochemical processes occurring at plant roots and substrate such as adsorption and sedimentation are the main driver for suspended solids (Tanner et al., 1995), phosphorous (P) (Arias and Brix, 2005) and heavy metals (Khan et al., 2009). Plant uptake of nutrients is also considered a major removal mechanism in CWs (Mesquita et al., 2013). Reduction of microbial pollutants, including pathogens and parasites, is determined by sedimentation, filtration at roots and substrate, predation and sunlight inactivation in open water areas (Jasper et al., 2013).

Wetland substrate is a porous particulate packed bed filtration medium that creates the body of a CW. The substrate occupies the largest proportion of a CW and plays a central role in the purification process and the stability of the system by providing physical support for wetlands plants (Wu et al., 2015b). Biofilm forming microbial communities in CWs are strongly influenced by both the substrate type and its topography (Meng et al., 2014). The substrate supplies adsorption sites for contaminants and many biological and chemical processes take place within its matrix (Calheiros et al., 2009). Therefore, careful substrate selection is critical for optimized wetland performance, while price and local availability have to be taken into consideration (Ballantine and Tanner, 2010). Location and depth of the substrate varies with the type of wetland. For vertical flow CWs the depth of the substrate ranges between 50-60 cm (Prochaska and Zouboulis, 2009). The top 10-20 cm facilitate aerobic microbial activity and subsequent biodegradation, while the remaining 40-50 cm of the filtration depth contribute to anaerobic removal of the chemical oxygen demand (COD) and total nitrogen (TN) as well as phosphorus adsorption (Tietz et al., 2007). The filtration bed can consist of substrate layers of different granular sizes that increase towards the bottom drainage layer (Ávila et al., 2015), and may include an additional organic substrate such as wood mulch (Myszograj and Bydałek, 2016) or biochar (Zhou et al., 2017). In the case of horizontal flow CWs, the effective substrate depth is between 25-60 cm (Carballeira et al., 2017). Horizontal flow CWs can employ both mineral and organic substrate materials (Andreo-Martínez et al., 2017), however, the multilayer substrate composition is less common compared to vertical flow CWs. In free surface water flow CWs it is common design practice to use 20-30 cm of rooting soil. However, in this type of wetland the substrate is considered to be of secondary importance (Vymazal, 2013a). For all types of CWs, small, round, evenly sized grains are most commonly used to fill the bed and average substrate diameters range from 3 to 32 mm (Tilley, 2014).

Wetland substrates can be divided into natural and manufactured materials, which also include recycled and industrial by-products (Ballantine and Tanner, 2010; Johansson Westholm, 2006; Wu et al., 2015a). Natural substrates such as soil, sand, gravel and marine sediments have been traditionally used as filter materials in CWs. These substrates are widely available and require little pre-treatment prior to application (Healy et al., 2007). However, clogging, poor adsorption capacity, low hydraulic conductivity and accumulation of contaminants such as heavy metals are common problems associated with these substrates (Johansson Westholm, 2006). More recently, both natural inorganic minerals such as anthracite, apatite, bauxite, calcite and zeolite (Molle et al., 2011; Seo et al., 2008; Stefanakis et al., 2009) and organic materials such as biochar, rice straw, peat, and wood mulch became established as CW substrates (Gupta et al., 2015; Kizito et al., 2015; Xiong and Mahmood, 2010). Recycled materials and by-products from mining, metal-making, construction, manufacturing and agriculture have also been used as substrate materials in CWs, ~~not only for their competitive pollutant removal efficiency but also to reduce waste disposal into the environment (Hu et al., 2012).~~ Recycled materials include alum sludge, coal and steel slag, fly ash, polyethylene plastic, oyster shells, tire chips, and construction waste such as bricks (Blanco et al., 2016; Chyan et al., 2013; Shi et al., 2017; Tatoulis et al., 2017). Some substrates have been specifically modified to improve treatment performance, mainly for better P removal (Ballantine and Tanner, 2010; Johansson, 1997) but also for improved ammonium (Zhang et al., 2013) and heavy metal removal (Lian et al., 2013).

Recent reviews have compared the role of different substrates on the removal of nutrients, including P removal of both natural and manufactured substrates (Vohla et al., 2011) and summarizing the structural differences and inherent properties of unconventional substrates such as zeolite, rice husk, alum sludge among many others, and their capacity for substantial N and organics removal from wastewater under optimized operating conditions (Saeed and Sun, 2012). Various substrates have been classified based on their ion-exchanging, P sorbing and electron donating properties (Yang et al., 2018). In general, substrates rich in mineral oxides of calcium (Ca), aluminum (Al), and iron (Fe) such as limestone, biotite, muscovite, steel slag, light weight expanded clays aggregates (LECA) have high capacities of P and N removal (Johansson Westholm, 2006), while organic substrates such as rice straw, compost, and wood mulches can be utilized by microbes as electron donors and thus enhance nitrification and denitrification processes (Cao et al., 2016). Specific studies have focused on substrates with extensive capacity for P removal such as clay bricks, fly ash, wollastonite, slag material, bauxite, shale, burnt oil shale, limestone, zeolite and LECA (Drizo et al., 1999; Johansson Westholm, 2006; Lima et al., 2018).

This review focusses on suitability of LECA as a substrate in CWs. ~~LECA is a manufactured substrate made of natural clay or other materials such as shale, apatite material and industrial by-products. LECA is made by burning the ingredients i.e. clay, at high temperatures in a rotary kiln. Final products are expanded pellets with many semi-closed pores that account for up to 90% of the particle volume. These pores are formed as a result of gas generated from combustion of organic components of the clay and water evaporation (Arioz et al., 2007). The first use of LECA as CW substrate was reported in the early 1990s (Jenssen et al., 1991). Since then, LECA has been extensively investigated as a substrate for CWs worldwide (Białowiec et al., 2011; Jenssen and Krogstad, 2003; Lima et~~

al., 2018; Nurk et al., 2009; Zhu et al., 1997). The increasing use of LECA in CWs has been attributed to its superior performance to remove P, N, heavy metals and organic compounds (Murray, 2000; Sposito et al., 1999; Zhou and Keeling, 2013). Based on its raw materials, LECA consists of minerals such as hydrous aluminum silicates, Fe, Mg and other alkaline minerals that are critical for binding ions (Bernhardt et al., 2014). For example, LECA with an estimated surface areas >3 $m^2 \cdot g^{-1}$ (Nkansah et al., 2012; Tabase et al., 2013), pore sizes in the range of 1-5 µm and an estimated porosity of 50-80% provided numerous sites for adsorption of pollutants (Bogas et al., 2012; Bonabi et al., 2014; Meng et al., 2015; Nawel et al., 2017). LECA has a water absorption capacity between 5-25% (Bogas et al., 2012; Castro et al., 2011; Nepomuceno et al., 2018) and the cation exchange capacity of LECA is estimated in the range of 9.5 $cmol \cdot kg^{-1}$ (Drizo et al., 1999). Coarse grain LECA enhances hydraulic conductivity while finer LECA with high surface area to volume ratio allows effective biofilm adhesion and microbial growth which in turn contributes significantly to biodegradation processes (Albuquerque et al., 2009; Białowiec et al., 2011; Brix et al., 2001).

This review specifically summarizes the current knowledge of LECA application in CWs design for wastewater treatment and its performance for a broad range of pollutants. The paper further examines the technical aspects of LECA incorporation into CWs design solutions with a wider attention to the importance and possibilities of LECA structural modifications enhancing the removal of different types of pollutants using CW technology.

Moreover, the review aims to shed light on the environmental concerns of LECA recycling and energy consumption.

1. Light Expanded Clay Aggregates (LECA)

1.1. Production, use and composition

LECA is a subtype of light weight aggregates (LWA), which is a heterogeneous group of low-density materials used for various civil engineering purposes (Mladenovič et al., 2004). LECA is marketed worldwide under commercial trademarks such as Filtralite® produced in Norway, Danish Leca®, Swedish LECATM, and the German LiaporTM. In the United States, LECA is produced under Stalite, Gravelite and Go Green commercial trademarks (Baker et al., 2014). LECA is foremost designed for construction purposes hence the manufacturing process aims to deliver a product with a strong but low density, porous, sintered ceramic core, a dense external surface to avoid water adsorption and a near-spherical shape to improve fresh concrete properties (Cheeseman et al., 2005). The usage of LECA-like materials for construction purposes is traced back to ancient civilizations as Sumerians, Greek and Romans (Chandra and Berntsson, 2002). Owing to its lightweight and thermal properties, LECA is used as component for thermal insulation concretes (Al-Jabri et al., 2005). The mechanical properties and structural performance of LECA have been also utilized for modern megastructures and high rise buildings, retaining walls, backfill of building and bridge supports (Holm and Valsangkar, 1993; Real et al., 2016). Due to its mechanical properties such as a high strength to weight ratio, its thermal features, and its good performance as rhizosphere substrate, LECA has been increasingly applied in storm water management schemes based on urban green infrastructure including green roofs, green walls, permeable pavements and thermal insulation concretes (Karami et al., 2018; Molineux et al., 2016; Pradhan et al., 2018; Sailor and Hagos, 2011; Sengul et al., 2011).

Traditionally, montmorillonite or illite types of clay are used as a raw materials for LECA production (Nkansah et al., 2012). More recently a wider range of natural and artificial compounds such as shale, apatite minerals, industrial by-products including coal or solid waste incineration, fly ash among others, have been integrated with clay to produce modified LECA (Ayati et al., 2018; Cheeseman et al., 2005; Molle et al., 2011). In addition, waste materials such as wastewater sludge (González-Corrochano et al., 2009), heavy metals contaminated soils (Ayati et al., 2018; González-Corrochano et al., 2014), granite and marble mining residues have been successfully incorporated into LECA (Moreno-Maroto et al., 2017a). However, mixing additives into LECA raw material is not a common practice, mostly due to practical constraints which favour the production of homogenous aggregates made of locally available raw material. Additionally, there is health and environmental concerns since industrial by-products could contain toxic substances, particularly heavy metals.

As a raw material, clay is widely available and affordable. Clay contains ample amounts of mineral oxides such as Fe, Mg, Ca and Al oxides (Grim, 1962). LECA produced from clays is manufactured by burning wet-formed granules at high temperatures ranging from 1000-1300 °C in a rotary kiln. In the oven the clay expands rapidly due to gas generating combustion of organic matter, pore water evaporation, thermal decomposition of carbonates and ferric oxides (Ayati et al., 2018). Pellet expansion can also be enhanced by addition of mineral oil which acts as an expansion agent in the process (Fakhfakh et al., 2007; González-Corrochano et al., 2009). The usage of other combustible additives (e.g. sawdust and chopped straw) has also been reported to increase the porosity with no impact on the specific surface area of the pellets (Dabare and Svinka, 2013). Physical properties of LECA (e.g. strength, density, and expansion behavior) could be further changed through a mixture of clay and powdered sodium carbonate (Na_2CO_3), quartz (SiO_2), iron (III) oxide as hematite (Fe_2O_3), or elemental iron (Fe) (Bernhardt et al., 2014). Besides the raw materials mineral composition, temperature regime during production determines the final properties of LECA. An increase in temperature and exposure time of the clay feed causes higher shrinkage of granules and results in a higher density and lower porosity, whereas lower temperature and shorter burning time has the opposite effect (Moreno-Maroto et al., 2017b).

LECA is commercially available in two forms: granular (intact) or crushed (Figure *3*). Geotechnical and construction applications predominantly use the intact specimens, while crushed LECA is used in hydroponics and water filtration applications (Bahmanpour et al., 2017). The LECA manufacturing process creates a pellet ranging from <1-32 mm with an average dry bulk density of about 400-600 kg m^{-3} and a smooth sintered ceramic outer shell that encloses the inner honeycomb structure (Ardakani and Yazdani, 2014; Musa et al., 2016). While LECA was initially designed as a light-weight geotechnical material for water retention and thermal isolation purposes its manufacturing process has not been optimized for applications that require a rather porous and sorbent surface as desired for CWs e.g. for P removal or as a matrix for biofilm growth. However, by crushing LECA granules, the interior porous structure is exposed to contact. Crushed LECA could have more than twice the specific surface area (1-10 m^2g^{-1}) compared to spherical LECA and a two times higher cation exchange capacity from 2.40 to 5.27 $cmol \cdot kg^{-1}$ (Kalhori et al., 2013; Stevik et al., 1999). Therefore, without modifying the manufacture process and raw material composition, it is possible to significantly improve the effectiveness of LECA materials by only crushing the granules to unlock their interior surface.

The chemical composition of LECA mainly depends on the mineralogy of its raw clay material. Clay minerals are hydrous aluminium silicates with Fe, Mg and other alkaline and earth alkaline metals at variable amounts (Uddin, 2017).

In terms of P removal, high Al content is more preferable than Fe, since Al is not redox-sensitive and is able to retain the adsorption capacity at low redox potential. In CWs, changes in redox potential could be imposed by the fluctuations of oxygen level resulting in water level changes or intense microbial activity and anoxic conditions promote Fe^{3+} reduction into Fe^{2+} which subsequently results in release of Fe bound P (Yaghi and Hartikainen, 2013).

The XRD analysis of clay materials used for LECA manufacturing typically shows the presence of quartz, alumina, hematite, and clay minerals. The exact elemental composition varies greatly as various classes of clays are used for LECA production including smectites, mica, kaolinite, serpentine, pyrophyllite, vermiculite and sepiolite (Shichi and Takagi, 2000). Different mineral composition is also accompanied with different pellet size distribution. During the treatment process, the pellets undergo a series of chemical changes e.g. decomposition of calcite and dolomite and CO_2 release followed by CaO formation. Therefore, the final product obtained after the firing process and expansion has a slightly different chemical composition than the raw material, missing hydrated mineral forms and organic content. LECA composition is generally dominated by 5-6 major constituents; 60-70% SiO_2, 15-18% Al_2O_3, 4-7% Fe_2O_3, 1-4% MgO, CaO, Na_2O, other constituents contributing less than 1% (Table 4).

2. Performance of LECA in CWs

2.1. Modified LECA materials

Comment [WU1]: Here the text was reduced from 793 to 670. Some sentences were rewritten to adapt the new text. minor deletion has no highlight.

The sorption process in CWs is a finite process, that requires periodic exchange of the wetland substrate (Arias and Brix, 2005; Drizo et al., 2002). Efforts to extend CW sorption performance have mainly focused on testing different substrates for better P removal efficiency. ~~P removal is central task in many treatment scenarios and therefore P is usually the critical sorbate that determines substrate saturation (Seo et al., 2005; Drizo et al., 2002). Moreover, measurements to manage clogging issues were also addressed in the~~

literature including preventative and restorative measures. Preventative measures to avoid clogging are mostly related to adjustment of hydraulic operation conditions and application of best management practices, while restorative measures may include treatment with chemicals or excavation and replacement of clogged substrates (De la Varga et al., 2013; Knowles et al., 2011; Lianfang et al., 2009; Nivala and Rousseau, 2009; Pedescoll et al., 2009).

LECA pellets may become either saturated with a sorbate or jammed by the accumulation of organic matter and sediments on the pellets surfaces which hinders LECA functionality and reduces its adsorption capacity (Ballantine and Tanner, 2010). Another option to increase the lifetime of a substrate is to alter its properties or add functionality. Several studies attempted to improve LECA through changing its specific mineral content or alter its surface charges via coating or use of additives such as dolomite and lime (Table 1). Coating LECA with Al and Fe, has a positive effect on LECA capacity for P and As removal from groundwater (Haque et al., 2008; Yaghi, 2015; Yaghi and Hartikainen, 2018, 2013). In addition, use of MgO nanoparticles as coating material has increased the LECA surface area and its adsorption capacity for removal of pharmaceutical pollutants (Kalhori et al., 2017).

The photocatalyst titanium dioxide (TiO_2) has been investigated as potential LECA coating for ammonia removal through photo-degradation under solar light (Shavisi et al., 2014; Zendehzaban et al., 2013) and UV irradiation (Mohammadi et al., 2016). The results of these studies presented high ammonium removal efficiency with the highest removal value (95.2%) achieved under UV radiation. TiO_2 sol-gel coating on LECA pellets was further investigated for antibiotics removal from wastewater, the resulting material was mechanically stable, had an enhanced adsorption capacity and was photocatalytic active (Pronina et al., 2015). Sohrabi and Akhlaghian (2016) applied copper-modified TiO_2 and iron-modified TiO_2 photocatalysts on LECA, the results indicated that copper modified LECA showed the best photocatalytic performance using phenol as a model pollutant.

Hydrogen-peroxide (H_2O_2) and magnesium chloride ($MgCl_2$) modified LECA were compared with unmodified LECA in their adsorption capacity for fluoride removal. The adsorption capacity of modified LECA increased roughly 2-3 fold, the achieved values were ~~8.53 mg g^{-1}, 17.83 mg g^{-1}, and 23.86 mg g^{-1} for natural LECA,~~ hydrogen peroxide modified LECA, and magnesium chloride modified LECA, respectively. The results were attributed to the positive charge of the oxide surfaces that were influenced by increasing pH values and the formation of fluoride ion complexes e.g. calcium fluoride which increased the ions adsorbed to LECA surfaces (Sepehr et al., 2014). Mixing of sodium carbonate (Na_2CO_3), silicon dioxide (SiO_2) and iron oxide (Fe_2O_3) at 2-10 wt% into the raw material, i.e. clay powder, was found to increase particle density, porosity and the crushing strength of LECA (Bernhardt et al., 2014). Lime had a positive effect on P adsorption capacity (Johansson, 1997). Mixing raw materials with fly ash and dolomite were found to enhance P and N removal capacity, hydraulic conductivity, and porosity (Białowiec et al., 2011; Jenssen and Krogstad, 2003). Other additives may alter the internal structure of LECA in order to obtain a more reactive surface. For example, the raw material for LECA can be enriched with mineral additives such as quartz sand to improve the particle size distribution and refine gas release during the expansion process (Fakhfakh et al., 2007).

Few studies have investigated the use of bioaugmentation technology to enhance denitrification and pollutants removal in LECA based CWs. The studies argued that LECA is a sterile substrate due to its exposure to high temperature during the manufacturing phase, and the microbes received by the influent are insufficient to carry on the process effectively, therefore introducing an already adapted microbial culture to a newly established CWs could positively influence nutrient removal processes (Nurk et al., 2009; Zaytsev et al., 2011) and perhaps lead to a faster achievement of treatment goals. Pinto et al. (2016) found that use of white-rot fungus *Lentinula edodes* to inoculate LECA and other substrates including cork; cork and straw and coat pine enhanced pesticide degradation by almost 50%. Bioaugmentation has been researched for many years for wastewater treatment application. However, results show that performance is rather difficult to predict compared to the earlier mentioned chemical and physical modification strategies (Herrero and Stuckey, 2015).

2.2. Pollutants removal through adsorption

Naturally occurring clay and clay minerals play important and complex roles in soil chemistry and its nutrient balance (Bohn et al., 2002). For example, clay minerals are involved in phosphate fixation (Gérard, 2016), heavy metal binding (Mercier and Pinnavaia, 1998), nitrate retention and ion-exchange (Mohsenipour et al., 2015). ~~Clays~~

~~also interact with the soil organic matter (Six et al., 2004) and microorganisms (Chenu et al., 2002).~~ Powdery or granular clay has been widely used as a low-cost locally available sorbent for water contaminants including, N, P, and heavy metals (Celis et al., 2000; Mena-Duran et al., 2007), arsenic (Lenoble et al., 2002), fluoride (Karthikeyan et al., 2005) and biocides (Lezehari et al., 2010).

The adsorption isotherms on clays in general can be described through several isotherm models. ~~The most widely used models for describing adsorption onto LECA are Langmuir and Freundlich (Vimonses et al., 2009) (Amiri et al., 2011; Dordio and Carvalho, 2013a; Sharifnia et al., 2016; Zhu et al., 2011). However, the Freundlich isotherm was found to fit better than the Langmuir isotherm (Sharifnia et al., 2016).~~

In LECA based CWs, especially in unplanted CWs, adsorption is one of the main routes for the removal of a wide range of water pollutants (Bahmanpour et al., 2017; Białowiec et al., 2009; Dordio and Carvalho, 2013a; Dordio et al., 2007; Põldvere et al., 2009).

~~Adsorption mechanism of oxyanions occurs via anion exchange mechanism and ligand exchange mechanism (Yaghi and Hartikainen, 2018, 2013). During the anion exchange mechanism, ions received by the influent are exchanged with similar charged ions bound to the functional groups contained within a solid matrix i.e. LECA (Yang et al., 2018). In this type of physical adsorption the bonding consists of a water molecule located between the anion and the surface of the substrate. However, this electrostatic bonding is considered rather weak and reversible. On the other hand, ligand exchange mechanism does not depend on surface charge of the mineral and can occur on positively, negatively as well as~~

~~on neutrally charged surfaces and include formation of multiple bonds (Essington, 2015). In the ligand exchange mechanism, oxyanions such as phosphate replace aqua groups (H_2O) or hydroxyl groups (OH^-) on the Al and Fe oxide's surface and bind directly to the surfaces of these oxides (Penn and Camberato, 2019).~~

~~Ligand exchange mechanisms occur preferably under acidic conditions, not only due to the positive surface charge that is usually formed at low pH, but also because of the increasing protonation of the OH^- groups at the mineral oxide surface, leading to the formation of aqua groups that swap more readily with oxyanions than OH^- groups. Under alkaline conditions, the mineral oxide surface is negatively charged and occupied OH^- ions with which hinders the adsorption of oxyanions (Yaghi, 2015).~~

2.2.1. Nitrogen adsorption

Nitrate and ammonium are adsorbed to clay surfaces via an ion exchange mechanism (Balci and Dinçel, 2002; Hokkanen et al., 2014). ~~The adsorption rate of nitrate onto the clay surface is a rather rapid process conditioned by the availability of the anion exchangers and the saturation of the adsorbate, thus the rate of adsorption may reduce over time (Mohsenipour et al., 2015).~~ Previous studies emphasized the capacity of clay in removing N species from wastewater through adsorption processes ~~(Oliveira et al., 2003; Rožić et al., 2000; Witter and Lopez-Real, 1988). Porous materials such as sepiolite, slag, activated carbon (Öztürk and Bektaş, 2004) and zeolite (Zhan et al., 2011), have been applied for nitrate removal. Many other adsorbents such as activated carbon (Huang et~~

~~al., 2008), agricultural residues (Liu et al., 2010), biochar (Gupta et al., 2015), bentonite (Angar et al., 2017) were investigated for ammonium removal.~~ High ammonium adsorption rates occur when the ammonium concentration in the water increases (Vymazal, 2007). However, ammonium is bound loosely to clay surfaces and can be easily oxidized to nitrate when exposed to oxygen in case of periodically draining the sorbent (Kadlec et al., 2017; Sun et al., 2006). Despite promising results for N removal by LECA via adsorption this mechanism is poorly quantified in literature. Sharifnia et al. (2016) investigated ammonium adsorption to LECA and found the maximum monolayer coverage capacity of LECA was 0.255 mg ammonium g^{-1}. The adsorption capacity was highest between pH 6-7, and equilibrium concentration was reached after 150 min with rapid adsorption within the first 60 minutes.

2.2.2. Phosphate adsorption

Comment [WU2]: This text was corrected,

Phosphate is adsorbed as inner-sphere complex with the oxygen atom of phosphate bound directly to Al and Fe-oxides at the LECA surface (Kwon and Kubicki, 2004; Zheng et al., 2012). Inner-sphere complexes are considered strong and mostly irreversible (Yaghi, 2015). The initial Ca, Fe, Al, and Mg concentrations affect the amount of P adsorbed by LECA surfaces (Baker et al., 2014). Among these elements, Ca has the strongest correlation with P-sorption capacity (Zhu et al., 1997). Therefore, low P removal in some LECA-based CWs (Table 1) can be attributed to the low Ca content of the substrate

(Johansson, 1997). A positive correlation was found between P removal and the content of both CaO and Ca in substrates (Vohla et al., 2011).

The pH is a critical parameter that affects the fate of phosphorous in CWs. Higher pH values have a positive effect on P adsorption and precipitation (Vymazal, 2007). The highest P adsorption (800 mg kg^{-1}) by LECA was achieved at a highly alkaline pH of 12.3 according to Zhu et al. (1997), while only 72 hours were needed to reach the maximum adsorption capacity of the substrate. The P adsorption in CWs involves two steps according to Jenssen and Krogstad (2003). The first step of adsorption can be considered as a short term transition stage and mostly occurs at low P concentration. This step is barely affected by the CW operational regime, including the hydraulic rate and the retention time. The second sorption step can continue for weeks or months depending on substrate properties and P concentration. High P concentrations can depress pH and eventually the precipitation process of P. Jenssen and Krogstad suggested therefore a retention time of 4 weeks for an optimal P adsorption by LECA under cold climate conditions.

LECA can have a strong influence on pH values of the water within the CW itself, because of its high contents of Ca minerals (Białowiec et al., 2011). Põldvere et al. (2009) measured high pH values in the outflow of a LECA based hybrid CWs monitored for one year with an average range from 8.1 to 8.8 in the first 9 months and from 7.6 to 7.7 in the remaining three months. ~~The pH values in LECA beds can range from 4.0 to 9.5 (Mesquita et al., 2013). Previous studies indicated that an effective P removal in LECA based CWs occurs at~~

~~high pH value ranging from 10 to 12 (Jenssen and Krogstad, 2003; Zhu et al., 1997).~~ However, highly alkaline conditions can adversely affect the growth of microbial communities which is important for organic matter and N removal processes (Tietz et al., 2007).

LECA has a finite capacity to adsorb P because of its ceramic matrix resulting from the high production temperature, which makes it resistant for both mechanical and environmental changes, therefore it is unlikely that new adsorption sites will emerge or generate in contrast to soil matrixes (Jenssen and Krogstad, 2003). In addition, poorer than expected adsorption performance can be also explained by blocking of sorption sites due to biofilm build-up and accumulation of organic matter at the granules surface (Knowles et al., 2011).

LECA systems can retain P through precipitation and sedimentation reactions with Ca-rich particles. The precipitation mechanism is favored at higher pH values or in presence of dissolved Ca in wastewater which promote P precipitation as Ca- phosphates especially during the initial stages of the treatment process (Jenssen and Krogstad, 2003). However, as pH values and dissolved oxygen concentrations within body of the CW start to decrease, further P precipitation is inhibited. Despite the significant contribution of wetlands sediments to P removal from wastewater, this P sink is often not considered in LECA based CW (Braskerud, 2002; Mendes et al., 2018).

2.2.3. Adsorption of heavy metals and organic pollutants

Clays, in general, have good removal capacity for heavy metals due to their high cation exchange capacity (Ma and Eggleton, 1999). This gives a good indication that LECA as a clay-based material can also provide efficient treatment for water contaminated with heavy metals. LECA has been applied to remove high concentrations of Pb and Cd from industrial wastewater (Table 3) (Malakootian et al., 2009) as well as Pb and Cu from mining tailings (Scholz and Xu, 2002). Pharmaceuticals such as MCPA (4-chloro-2-methyphenoxyacetic acid), oxytetracycline, and polyphenol can be removed by electrostatic interactions which is partly driven by the extensive protonation of LECA surface at neutral pH values where these compounds are mostly in the anionic form while LECA surfaces are positively charged (Dordio and Carvalho, 2013b; Dordio et al., 2007). The LECA capacity for lipophilic (oxybenzone and triclosan) and hydrophilic compounds (caffeine) was also investigated. The results revealed higher removal of lipophilic compounds compared to hydrophilic compounds (Ferreira et al., 2017).

LECA was reported to remove polycyclic aromatic hydrocarbons (PAHS) including phenanthrene, fluoranthene and pyrene compounds (Nkansah et al., 2012) (Table 3).

The study attributed this removal to LECA’s exterior and interior surfaces that exhibit hydrophobic character, however, the underlying mechanisms are rather vaguely understood as factors that provide hydrophobic capacities to LECA are not well addressed in the existing literature. In addition, LECA made of clays may lack for hydrophobic characteristics as clays have weak adsorption capacity for hydrophobic compounds normally favored by the strong hydration capacity of their inorganic exchangeable ions (Acikyildiz et al., 2015).

2.3. Pollutants removal through biological pathways

2.3.1. Biological nitrogen removal

The main biological pathways for N removal in CW involve microbial degradation (Li et al., 2014) and both uptake and assimilation by plants and microorganisms (Wu et al., 2011). ~~The microbial degradation which takes place under aerobic and anaerobic conditions comprises three steps: ammonification, nitrification, and denitrification. In the first step, organic N is converted into ammonia in aerobic and anaerobic zones of the CWs. Ammonia is removed via the nitrification process under strict aerobic conditions by special types of nitrifiers such as *Nitropira, Nitrosococcus*, and *Nitrobacter* (Mayo and Bigambo, 2005). Denitrification is carried out by heterotrophic microorganisms that need organic matter to obtain their energy, the microorganisms under anoxic conditions use nitrate as terminal electron acceptor and organic C as electron donor to produce gaseous N. Each step of the microbial degradation can be greatly affected by environmental factors such as oxygen availability, water temperature, pH, organic matter and the presence of the specific microorganisms (Vymazal, 2007).~~ The nature of the CW substrate is a main factor determining the location and the activities of the microbial community (Truu et al., 2009). Previous studies have shown a decline in microbial density in the upper 10 cm of the substrate when porous materials as sand and gravel were used as filtration bed

(Braeckevelt et al., 2007; Nurk et al., 2005). The reallocation of the microbial biomass into greater depths can be explained by the higher availability of organic matter and the shelter provided on the substrate surfaces and within the micropores between LECA grains (Calheiros et al., 2009; Tietz et al., 2007). Many studies attributed LECA capacity for high N removal to its high porosity and large surface area (Saeed and Sun, 2012; Vymazal and Kröpfelová, 2009; Yang et al., 2018). High porosity allows additional oxygen to penetrate, especially if LECA is installed as an upper layer.

~~The selected vegetation plays an important role for wastewater treatment in CW systems, not only through uptake and assimilation of nutrients but also because~~ the plant roots provide surface area for biofilm formation and growth, and create aerobic zones that are important for microbial communities involved in the biological degradation (Allen et al., 2002). ~~Previous studies have shown that ambient oxygen release into the rhizosphere is supplied by macrophyte plant roots~~ (Brix, 1993; Gagnon et al., 2007; Wu et al., 2001). In vertical flow CWs, large proportion of the oxygen enters the substrate bed via diffusion, while in horizontal flow, the oxygen is mostly provided by the plants (Lee et al., 2009; Molle et al., 2006). Decaying roots provide readily accessible organic matter as additional carbon source and can remarkably ~~improve denitrification rates and thus improve N removal in CWs (Lu et al., 2009; Luo et al., 2018). The roots can also provide surface area for attached microbial growth (Clairmont et al., 2019)~~. Overall, planted LECA beds have been reported to have higher N removal capacity due to higher microbial diversity and density compared to unplanted ones (Almeida et al., 2017; Białowiec et al., 2009; Dordio and Carvalho, 2013a).

2.3.2. Biological removal of P

P uptake and assimilation by plants are the main biological routes for P removal in CWs (Kim and Geary, 2001). The largest proportion of soluble P is taken up by microphytes and algea, especially in the early stages of the growing season. P uptake by plants contributes to a short term removal mostly during growth (Vymazal, 2007) and if not removed decaying plants may lead to re-release of P into the wetland. Organic P which enters the CW as phospholipids, nucleic acids and sugar phosphates is transformed via the microbial metabolism. The microbial uptake of P is very fast and accounts for a temporary removal as microorganisms have a very short turnover rate (Qualls and Richardson, 2000). However, biological take-up of P in LECA based CW systems is not quantified due to the dominance of P is removal through adsorption.

2.4. Pathogens removal

CWs have been increasingly adopted for wastewater reuse schemes, therefore pathogen removal has become a central treatment goal that determines wetland design and operation (Barbagallo et al., 2010; Masi et al., 2007). ~~Current research targets mostly common microbial indicators for fecal contamination, such as *E. coli*, fecal streptococci, *C. perfringens*, or *Giardia lamblia* (Wu et al., 2016). However, other pathogenic~~

~~microorganisms such as Salmonella, polioviruses and *Cryptosporidium spp* have also been investigated (Redder et al., 2010; Sidhu et al., 2010).~~

CWs provide a number of biological, physical and chemical removal mechanisms for pathogens which mimic processes occurring in natural wetlands (Kadlec and Wallace, 2008). ~~Depending on the system's flow regime (surface or subsurface flow), pathogen inactivation is mostly~~ driven by a combination of sedimentation and filtration, adsorption, predation, photoinactivation, natural die-off as well as biocidal effect of root exudates or internalization into plant tissue (Alufasi et al., 2017; Boutilier et al., 2009; Wand et al., 2007; Wenk et al., 2019; Wu et al., 2016). ~~The effectiveness of given removal mechanisms might be enhanced through adequate hydraulic management (Giácoman-Vallejos et al., 2015), presence of specific vegetation (García et al., 2013), the wastewater influent composition (Yang et al., 2012), seasonal weather patterns (Morató et al., 2014) or aeration (Headley et al., 2013). Further factors affecting pathogen removal effectiveness include size and type of substrate media (López et al., 2019).~~ Filter media in CWs contribute mostly to physiochemical pathogen removal mechanisms such as filtration and adsorption. Fine granular substrates trap microorganisms and increases their retention time by enhancing removal through natural-die off (Vacca et al., 2005). Adsorption of pathogens was found to be particularly effective for substrates with positive surface charge (Rzhepishevska et al., 2013). Both chemical composition and physical substrate properties, for example porosity, affect the microbial composition and biofilm growth and contribute to pathogen predation and adhesion (Long et al., 2016; Meng et al., 2014)..

However, the link between substrate properties, predation and microbial composition in CWs is currently not fully understood. ~~Some substrate media, i.e. steel slag, cause pH variations leading to local and whole system acidification or alkalization which would impose additional stress on pathogen survival rates, yet this phenomenon is still poorly studied~~ (Lee et al., 2010; Mayes et al., 2009). CWs for primary and secondary wastewater treatment operate at average influent *E.* coli concentrations of 10^5-10^8 colony forming units per 100 mL (cfu/100mL) for domestic wastewater (Headley et al., 2013) and up to 10^{11} cfu/100mL for fecal coliforms in slaughterhouse wastewater (Rivera et al., 1997). The typical removal rates of fecal microorganism observed in CWs range from 1-3 log units (Abou-Elela et al., 2013; Headley et al., 2013; Molleda et al., 2008). Occasionally, removal above 3 log units was also recorded, both in single stage and hybrid systems (El-Khateeb et al., 2009; Pundsack et al., 2001). In terms of water quality standards for water reuse, the free water surface systems located in tropical or subtropical climates are capable of producing final effluent with fecal-coliform concentration as low as 100 cfu/100 mL (Greenway, 2005), while in temperate climates, the effluent could be consistently maintained around 1000 cfu/100 mL (Vivant et al., 2016). Subsurface flow systems may achieve effluent concentration below 1000 cfu/100ml, particularly when employed as tertiary treatment step (Adrados et al., 2018; Andreo-Martínez et al., 2017). Nevertheless, many CWs exhibit high variability in effluent pathogen concentrations, and further research is needed to improve design towards a more consistent removal performance (Jasper et al., 2013; Wenk et al., 2019).

Due to the coarse granular size (5-20 mm), the water in LECA filtration beds has a relatively low residence time in comparison with sand beds, therefore bacterial adhesion mechanisms may not be very effective (Ausland et al., 2002). Similarly, large granular size also excludes both filtration and straining from being an important removal mechanism in LECA-dominated systems (Díaz et al., 2010). On the other hand, LECA’s porous surface enhances biofilm growth and subsequent bio-clogging, which facilitates effective bacteria immobilization (Lianfang et al., 2009). The high cation exchange capacity of LECA could be also beneficial for bacterial removal since it enhances adhesion (Stevik et al., 1999). Additionally, clay minerals in LECA, may alter i.e. metabolic pathways of biofilm microorganisms encapsulating the granule through increase of cell division in *E. coli* in the presence of kaolinite (Cuadros, 2017). As a proven soilless plant growing substrate (Pradhan et al., 2018), LECA may facilitate pathogen removal through root biofilm attachment (VanKempen-Fryling and Camper, 2017) and possibly plant exudates (Alufasi et al., 2017).

Consistent *E.* coli removal of 1.5 log-units was reported for a LECA-based horizontal flow polishing CWs after a prior filtration step, and the removal performance was similar to gravel systems that were operated in parallel (Verlicchi et al., 2009). Removal rates of up to 3 log for *E. coli* and total coliforms were reported in horizontal flow LECA CW located in North Portugal planted with a polyculture of ornamental flowering plants (Calheiros et al., 2015). Paruch (2010) speculated that the integration of LECA-based CW with preceding septic tanks could completely eliminate the dissemination of human parasitic helminth eggs. LECA upflow biofilters designed as unplanted subsurface CW, showed full removal of somatic coliphages which was attributed to the extensive attraction of negatively charged viruses onto the positively charged LECA surface (Heistad et al., 2006). Due to the potential to reuse LECA as soil enhancer in agriculture, sanitation safety issues have been investigated. *E. coli* contamination of LECA from a horizontal flow CW-derived LECA persisted for more than 14 months after the last contact with wastewater (Paruch, 2011). However, despite the long survival time, *E. coli* concentrations below 2.5 10^3 cfu/g of dried substrate, allowed reuse for agricultural applications according to Norwegian legal requirements (Paruch et al., 2007). Survival of coliform bacteria on LECA has been further tested to assess the health hazards related to the use of vertical flow CW in densely populated areas (Bydałek and Myszograj, 2019). When exposed to atmospheric conditions as a top filtration layer in vertical flow CWs, LECA showed slower inactivation rates of coliforms (k_{6h}=0.36h^{-1}, k_{12h}=0.25h^{-1}) in comparison to gravel or slag but faster inactivation compared to organic substrates such as bark and charcoal.

2.5. Organic matter removal

The removal of organic matter i.e. BOD, COD and total suspended solids (TSS) in CWs is driven by microbial degradation and the retention of these compounds to the substrate bed (Saeed and Sun, 2012). LECA substrate has a good capacity for organic matter removal because of high porosity and specific surface areas which allow better biofilm adhesion to increase the biodegradation (Table 2). In a hybrid LECA CW, almost complete removal of BOD (99%) was achieved (Põldvere et al., 2009; Zaytsev et al., 2007). A high removal of

COD (92%) and TSS (80%) was also reported by Dordio and Carvalho (2013a) in CW mesocosms with more than 60% of the organic matter removed by sedimentation on the LECA bed. The sedimentation of the organic matter occurs mostly near the CW inlet (Caselles-Osorio et al., 2007). Organic matter accumulation is strongly correlated with organic loading rates (Meng et al., 2015).

The high average removal of both BOD (91%) and TSS (78%) in a vertical flow CW was attributed to the efficient mineralization of organic matter (Öövel et al., 2007). The removal of BOD, COD and TSS was found to be affected by the vegetation type and the creation of aerobic zones within the rhizosphere which positively affected microbial density and metabolism (Lima et al., 2018).

3. Design considerations for LECA-based CWs

3.1. Layout of CWs using LECA substrate

In the majority of CW designs the substrate is arranged into horizontal layers (Kadlec and Wallace, 2008). In larger more heterogeneous treatment wetlands with various sections or consecutive treatment cells different types of substrate may be used spatially (Lu et al., 2016). Simple design CWs contain a single substrate, that is usually confined by an impermeable bottom liner (Almeida et al., 2017) such a design is particularly common in decentralized, rural areas, where CWs serve single households (Figure *4*). Multi-layered wetlands have been constructed with up to three different layers, while double layers are most common (Vymazal, 2013b). Using double layers in vertical flow may create different oxic conditions as nitrifying bacteria prefer to attach to porous and well aerated media, whereas denitrifying bacteria colonize more compact aggregates that support low oxygen conditions (Białowiec et al., 2011). Multilayers can be exclusively composed of LECA granules of different grain sizes or incorporate different types of substrates (Białowiec et al., 2011; Calheiros et al., 2009). Horizontal positioning of different substrate layers is variable. LECA has been mostly used as the upper layer when applied with other substrates to remove suspended solids and promote the growth of nitrifying microorganisms while providing aeration (Almeida et al., 2017). On the other hand, installing LECA as a bottom layer substrate has a positive effect on the hydraulic conductivity and protects the system against clogging (Suliman et al., 2006). Layer arrangements uniformity and grain size distribution within each layer are also critical for adequate hydraulic conditions to minimize clogging issues (Brix et al., 2001). The grain sizes used in LECA beds can range from smaller 1 mm (powdery form) to 10/20 mm, sizes of 2/4, 3/8, 4/10 and 13/15 mm have also been installed for different types of CWs (See table 2). Different depths of LECA layers were tested to compare performance with thicknesses ranging from 12 cm to 150 cm in lab trials using columns or mesocosms with narrow volumes e.g. 0.25 m^2 (Almeida et al., 2017; Białowiec et al., 2011; Nurk et al., 2009; Özengin, 2016). LECA layer depths ranging from 20-90 cm have been used in a three layer hybrid CW of an area of 216 m^2 for domestic wastewater treatment. For the vertical flow section of this wetland a layer of 50 cm of coarser granules 10-20 mm was used as bottom layer covered by 30 cm of finer 2-4 mm granule to ensure oxygen transport. The vertical bed was followed by a horizontal subsurface flow filter (90 cm in depth), filled with 2-4 mm LECA granules (Öövel et al., 2007). Põldvere et al. (2009) installed three layers of LECA with 25 cm, 20 cm and 20 cm thickness of bottom, middle and top layer, respectively, in a 70 cm deep vertical filter, coarser granules of 10–20 mm were used as a bottom layer to maintain hydraulic conductivity.

3.2. LECA CWs for different types of wastewater

Constructed wetlands have been used for the treatment of a wide range of different types of water including domestic, agricultural and industrial sources (Vymazal, 2009). Both agricultural and industrial wastewater may exhibit high loads of certain contaminants and contaminant classes, which requires case by case CW design considerations. For example, dairy farm and aquaculture effluent can be high in COD, proteins, N species and phosphate (Dauda et al., 2019; Justino et al., 2016; Nagarajan et al., 2019), and greenhouse effluent is usually high in nitrate (Prystay and Lo, 2001). The composition of domestic wastewater is usually more similar across different locations (Tran et al., 2015). Typical values of main wastewater parameters to size CWs were proposed by Kadlec and Wallace (2008): BOD 220 mg l^{-1}; TSS 500 mg l^{-1}; TN 40 mg l^{-1}; and P 8 mg l^{-1}.

Physiochemical properties of LECA (Figure *5*) allow for application in domestic wastewater treatment with the aim to remove N species, organic matter and P (Albuquerque et al., 2009; Lu et al., 2016; Meng et al., 2015; Özengin, 2016). For this type of wastewater LECA containing CWs have achieved a maximum reduction of 99% BOD, 94% TSS, 83-99%

ammonium and 89% P (Table 2). Organic matter removal in LECA based CWs is significant for all types of wastewater. LECA has shown a relatively good capacity for P removal from domestic and food processing wastewaters with values ranging from 60% to 67.3% (Özengin, 2016; Põldvere et al., 2009; Zaytsev et al., 2007). LECA substrate has been also applied to remove heavy metals from urban runoff and a wide range of industrial wastewater including mining tailings, tanneries and dye factories (Calheiros et al., 2008; Malakootian et al., 2009; Scholz and Xu, 2002) and agricultural wastewater include olive mill effluent and swine wastewater (Dordio and Carvalho, 2013a). Accumulation of organic matter and clogging at the inlet of CWs is a major challenge for high COD treatment tasks (Healy et al., 2007; Langergraber et al., 2003). Coarse LECA substrates ranging from 8-10 mm have been shown to facilitate clogging issues, while smaller sized LECA substrates of 1-4 mm could not prevent clogging efficiently (Albuquerque et al., 2009; Suliman et al., 2006). Pre-dilution of raw wastewater before being introduced to the CW coupled with using fine particles (2-4 mm) can minimize the clogging problem resulting from the accumulation of organic matter(Dordio and Carvalho, 2013a).

3.3. Hydraulic loading rate and hydraulic retention time

The hydraulic conditions such as retention time and loading rate are vital factors determining the treatment process in CWs (Ghosh and Gopal, 2010; Jing et al., 2002; Persson et al., 1999). The hydraulic loading rate should be balanced with the expected oxygen depletion along the wetland (Liu et al., 2016). Generally, low hydraulic loading rates and increasing hydraulic retention times lead to greater nutrient removal efficiency (Almeida et al., 2017), whereas organic overloading results in hydraulic dysfunctions via clogging (Knowles et al., 2011).

The effect of hydraulic loading rate and hydraulic retention time has been investigated in several LECA beds for P, N and organics removal. Herrmann et al. (2013) found that a loading rate of 100 L m^{-2} d^{-1} increased the average P binding capacity of LECA wastewater filters to 1.1 g kg^{-1} at residence times ranging from 5 to 15 min. High removal capacity of P in LECA beds is attributed to the hydraulic conductivity and the adaptability of LECA to changing hydraulic loads (Öövel et al., 2007). Effluent recirculation enhances nitrification processes through increasing both the contact time of wastewater with CW biofilms and the supply of oxygen and organic matter into the wetland (Saeed and Sun, 2012). Effluent recirculation has been tested for a hybrid LECA CWs, it was found that high recirculation rates of up to 300% in a hybrid CW can increase removal efficiency for BOD, TSS, total N (Table 2) (Põldvere et al., 2009; Zaytsev et al., 2007). A hydraulic loading rate of 239 ± 7 L $m^{-2}d^{-1}$ at a hydraulic retention time of 140 min was found to increase nitrate removal by maximum 66%, any further increase in hydraulic loading rate was found to have an opposite result on nitrate removal rate (Almeida et al., 2017). Dordio and Carvalho (2013a) indicated that LECA adsorption capacity in planted beds was most effective after 6 days for TSS (95.3%), and COD (92.5%) and 9 days for ammonium (75.2%) and nitrate (58.4%).

3.4. Dissolved oxygen

Oxygen supply drives the metabolic processes responsible for BOD/COD removal and nitrification (Ding et al., 2012). Oxygen transfer rates and horizontal dissolved oxygen (DO) transport into CWs are determined by the type of wastewater, the wetland depth, the vegetation and the substrate (Vymazal and Kröpfelová, 2008). In planted CWs oxygen diffusion and oxygen release by macrophyte roots are the major routes of oxygen transport (Li et al., 2014; Vymazal and Kröpfelová, 2008). The oxygen concentration of influent wastewater can range from almost anoxic (0.6 mg l^{-1}) to almost saturated (7.8 mg l^{-1}) levels (Liu et al., 2016). Complete oxygen depletion in CWs is nevertheless common when treating high organic or N loaded wastewaters (Albuquerque et al., 2009). The depth of the filtration bed influences DO distribution within CWs. As depth increases, there is more volume available for microbial degradation processes (García et al., 2004) while shallow beds have a larger air-water interface allowing better oxygen transfer than deeper beds (Kadlec et al., 2017). In vertical flow CWs more than 90% of the oxygen penetrates the system by air diffusion; most of it is consumed by COD removal and nitrification processes in the upper zone (Li et al., 2014).

Porous, large grained and loose substrates enhance oxygen transfer into the filtration bed (Verhoeven and Meuleman, 1999), although LECA is a porous substrate, low DO concentrations have been an issue similar to other types of substrates (Mesquita et al., 2013).

The reported values are ranging from 0.5 mg l^{-1} to 1.5 mg L^{-1} (Albuquerque et al., 2009; Lima et al., 2018) which is the minimum DO concentration required for nitrification is. Many studies indicated that shorter hydraulic retention time ranging from hours to a few days can create favorable conditions for efficient use of oxygen by the microbial biomass. High DO fluxes may eliminate the anoxic conditions inside the substrate and result in weak denitrification (Shuib et al., 2011; Tao et al., 2006; Xiao et al., 2010). Solutions to improve oxygen transfer into CWs, include optimization of vegetation and hydraulic conditions in addition to active aeration (Liu et al., 2016; Ouellet-Plamondon et al., 2006). In LECA based CWs recirculation of the effluent back to the influent can improve aeration conditions and overall purification efficiency (Põldvere et al., 2009) such as BOD removal (Zaytsev et al., 2007). Alternatively, batch (drain and fill) feed mode can create more oxygen-rich conditions compared to continuous feed mode, and increase N, P and COD removal (Zhang et al., 2012).

3.5. LECA CWs under different climatic conditions

CWs have been operated under a variety of climate conditions (Jenssen et al., 2005; Koottatep et al., 2005; Quanrud et al., 2004). Cold climate can significantly affect hydraulic performance and both biological and chemical processes in CWs; microbial activity and vegetation growth are reduced at low temperatures (Werker et al., 2002). The N removal is reported to be inhibited below 10 °C (Luo et al., 2005) and nitrification does not occur below 4°C (Cookson et al., 2002). A decrease in water temperatures from 20 to 5°C was found to decrease the adsorption capacity of LECA by 24 to 64%, increasing with grain size (Zhu et al., 1997). Several design alternations were implemented to improve wetland performance in cold climates. Lower hydraulic loading and both selection of tolerant vegetation and adapted substrates were found to increase the treatment performance (Yan and Xu, 2014). LECA has been extensively used for CWs in cold climate (Brix et al., 2001; Jenssen et al., 2005; Johansson, 1997; Mæhlum, 1995; Suliman et al., 2006) and in subtropical climates. The use of CWs in arid and semiarid environments in particular the Middle East and North Africa (MENA) region, is rather new, ~~despite the need for water treatment given population growth along with rising wastewater discharge volumes (Almuktar et al., 2018; Zidan et al., 2015). Wastewater treatment and reuse in the MENA region are challenged by inadequate technical knowledge as well as financial, logistic, and cultural constraints (Qadir et al., 2010).~~

4. Recycling of wetland substrates and environmental concerns

CWs have become an accepted and established technology for the treatment of water. More recently concerns have been raised about the fate of the substrates after the end of their useful lifetime (Yang et al., 2018). Substrates upon saturation may contain high concentrations of nutrients, organic compounds and in some cases, toxic contaminants and pathogens (Hench et al., 2003). The fate of the wetlands substrates after saturation is rather vague and poorly addressed in the literature (Jenssen and Krogstad, 2003; Johansson Westholm, 2006). Many studies highlighted the possibility of using spent LECA from CWs as P fertilizer and soil liming amendment for acidic soils (Jenssen et al., 2010; Johansson Westholm, 2006; Vohla et al., 2011) considering LECA's P adsorption potential which can reach up to 12,000 mg P kg^{-1} (Ádám et al., 2007; Ádám et al., 2006). However, P saturated LECA may not support short term P release in soils, including availability to plants. Hylander and Simán (2001) tested different types of saturated substrates with barley plants, and found that P-saturated LECA resulted in lower yields compared to crystalline slag substrates. In LECA P was bound tightly to Al and Fe oxides, while the P in slag was bound to Ca and was more readily available for the plants.

Production of LECA is known to have a high energy demand (Johansson Westholm, 2006), but actual quantitative information is virtually absent in literature, we only found one website based reference. This data indicated that amount of energy needed for producing 1 m^3 of LECA was estimated to be 931 MJ, while the CO_2 emission potential was 54 kg for the same quantity (www.leca.com). Therefore LECA is considered a high energy consumption manufactured substrates, its costs are determined by the production process rather than by the raw materials (Ballantine and Tanner, 2010). Sustainable solutions for recycling and regeneration of LECA are needed to manage its fate and minimize energy consumption.

5. Future research directions

LECA is an adsorptive material that has a high removal capacity for Phosphorus (P) compared to other types of constructed wetland substrates. Beyond P, interactions of

LECA with wastewater contaminants including organic trace contaminants, certain pathogens, in particular viruses, but also the nitrogen (N) species ammonium and nitrate need further investigation. Although, N removal in constructed wetlands occurs mainly through biological routes, substrates such as LECA may provide a buffer capacity, when metabolic processes temporary slowdown. Modification to tailor LECA for specific use in constructed wetland applications for better performance of desired treatment tasks or to improve biofilm development, including addressing clogging issues has untapped potential. Such modified properties might be achieved through relatively simple means by crushing pellets to expose the inner structures or by blending additives into raw clay mixtures. There is need to develop reuse and recycling strategies for spent constructed wetland substrates, including opportunities for P recovery, while considering potential heavy metals and pathogen loads. The energy required during LECA production needs to be accounted for when assessing its life cycle in comparison with alternative substrates.

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Tables

Table 1. Coatings and additives used for LECA properties and adsorption capacity improvement.

Coatings/ Additives	Treatment	Effects	Reference
Fe and Al oxides	As in groundwater	High adsorption capacity for As ions obtained at pH 2, 4 and 6.	Yaghi and Hartikainen (2018)
Fe oxide	As in groundwater	Faster adsorption, including increased capacity. Maximum As accumulation 3.31 mg of g^{-1} LECA at pH 6 to 7.	Haque et al. (2008)
Fe and Al oxides	P in groundwater	High adsorption capacity. Al-coated sorbents were superior to Fe-coated ones.	Yaghi and Hartikainen (2013)
MgO nanoparticles	Pharmaceuticals: metronidazole antibiotic	High specific surface area (76.12 m^2/g). Antibiotic adsorption increased by approximately 33% as adsorption sites increased.	Kalhori et al. (2017)
TiO_2 photocatalyst	Ammonia	High removal efficiency. The maximum degradation of NH_3 occurred at pH 11.	Zendehzaban et al. (2013); Shavisi et al. (2014)
TiO_2 sol-gel photocatalyst	Pharmaceuticals: tetracycline antibiotics and doxycycline	Improved mechanical stability Satisfactory photocatalytic antibiotic oxidation efficiency.	Pronina et al. (2015)
Fe/TiO_2 and Cu/TiO_2 photocatalysts	Phenol from synthetic wastewater	61 % degradation of phenol in synthetic wastewater.	Sohrabi and Akhlaghian (2016)

TiO_2 /Zinc oxide (ZnO)/LECA hybrid photocatalyst	Ammonia from synthetic wastewater	95.2% of ammonia removal during the first 3 hours.	Mohammadi et al. (2016)
H_2O_2-modified LECA	Water contaminated with fluoride	Increased surface area and adsorption capacity.	Sepehr et al. (2014)
$MgCl_2$-modified LECA		Fluoride adsorption capacities of 17.83 mg/g and 23.86 mg/g for H_2O_2-modified LECA and $MgCl_2$-modified LECA respectively.	
Na_2CO_3 SiO_2 Fe_2O_3	Physical characteristics of LECA	Decreased viscosity of the surface. No effect. Pore size increased and density reduced.	
LECA made of fly ash from sewage sludge		Promoted activity of a consortium of micro-organisms responsible for N removal. Provided loose, porous, and well-aerated substrate thus nitrifying bacteria prefer to attach to it. Increased N removal efficiency.	Białowiec et al. (2011)
$CaCO_3$/ Lime	P removal capacity	P adsorption increased	Johansson (1997)
Dolomite	N, P removal capacity and physical characteristics	High N and P removal. Enhanced LECA hydraulic conductivity, porosity and its insulation properties.	Jenssen and Krogstad (2003)
Quartz sand (< 250 µm grain size) and 1% motor oil (expansion promotor)		Better expansion properties. Physical properties such as apparent density and mechanical resistance improved.	Fakhfakh et al. (2007)

	Biological additives		
Bioaugmentation in a newly established LECA-based horizontal flow	Biochemical process	Change in the structure of the microbial community. High performance and stable denitrification process.	Nurk et al. (2009); Zaytsev et al. (2011)
Bioaugmentation using white-rot fungus *Lentinula edodes*	Pesticides group: terbuthylazine, difenoconazole, diflufenican and pendimethalin.	Moderate retention capacity of pesticides. Microbial activity enhanced by porosity.	Pinto et al. (2016)

Table 2. The removal efficiency of LECA substrates integrated with different types of CWs for N, P and organic compounds from diverse types of wastewater

LECA/ other substrates	wastewater source	CW type	Planted/unplanted	Total N		NH4-N[1]		NO3-N[2]		Total P		TSS[3]		BOD[4]		COD[5]		HLR	HRT[6] (d)	Reference
				In	Out	In	Out	In	Out	In	Out	In	Out	In	Out	In	Out			
				$mg\ l^{-1}$	%	$mg\ l^{-1}$	%	$mg\ l^{-1}$	%	$mg\ l^{-1}$	%	$mg\ l^{-1}$	%	$mg\ l^{-1}$	%	$mg\ l^{-1}$	%			
Bottom layer: 10 cm, LECA 10-20 mm. Middle layer: 25 cm, LECA 2-4 mm. Top layer: 10 cm LECA 3-8 mm	olive mill wastewater	vertical	planted	-	-	-	-	-	-	-	-	616	95	-	-	2160	92	-	6	Dordio and Carvalho (2013a)
			Unplanted	-	-	-	-	-	-	-	-		95	-	-		81	-		
Bottom layer: 10 cm, LECA 10-20 mm. Middle layer: 25 cm, LECA 2-4 mm. Top layer: 10 cm	swine wastewater		planted	-	-	392	75.2	24	58.4	-	-	480	86	-	-	1420	80	-	9	
			unplanted	-	-		47.4		52.3	-	-		86	-	-		68	-		

LECA 3-8 mm																				
20 cm LECA 13-15 mm	synthetic wastewater	sequencing batch mode	planted	69	19	40	-35	-	-	19	18	-	-	-	-	203	55	-	48, 72 h	Lima et al. (2018)
			unplanted		9		-32		-		25	-	-	-	-		47	-		
LECA	synthetic domestic wastewater	horizontal	planted	50	70	6	66	0.9	52	8	61	-	-	-	-	-	-	-	3	Özengin (2016)
			unplanted		65		57	0.9	66		67	-	-	-	-	-	-	-	-	
2-4, 4-10, 10- 20 mm	domestic sources and food processing plants	hybrid	unplanted	-	81	-	79	-	-	-	67	-	-	-	99	-	-	0.2-0.73 m^3 d^{-1}	-	Zaytsev et al. (2007)
Limestone LECA		vertical filled with crushed limestone and a horizontal filled with LECA		-	82	-	83	-	-	-	60				99	-				
LECA 2-4, 4-10, 10- 20 mm; limestone	secondary treatment of domestic wastewater	hybrid	unplanted	72	47	-	-	-	-	20	66	132	94	405	82	745	64	52 mm d^{-1}	6	Põldvere et al. (2009)
2–4 mm LECA	secondary treatment of domestic wastewater	batch mode	unplanted	54	82	-	-	-	-	6.6	48	33	82	135	99	224	70	59 mm d^{-1}	4	Põldvere et al. (2009)
FASSTT with LECA and gravel	artificial wastewater	vertical	planted	-	59	-	99	-	-	-	-	-	-	-	-	-	-	4.6 mm d^{-1}	7	Białowiec et al. (2011)
			unplanted	-	46	-	61–66	-	-	-	-	-	-	-	-	-	-			
4 to 8 mm	domestic wastewater	horizontal	unplanted	-	-	-	61-91	-	100	-	-	-	-	-	-	-	64-94	3.5 cm d^{-1}		Albuquerque et al. (2009)
2-4 mm	pretreated domestic wastewater	horizontal	unplanted	-	-	-	-	-	83	-	-	-	-	-	60	-	-	-	1-4,7	Nurk et al. (2009)
Filtralite® 4-8 mm	synthetic wastewater	horizontal	planted	-	-	36.3	59.3	-	-	-	-	-	-	-	-	315.9	74	3.6 cm d^{-1}	6	Mesquita et al. (2013)
			unplanted	-	-	26.7	33.9	-	-	-	-	-	-	-	-	311.2	38			

LECA 10/20	synthetic wastewater	vertical	planted	-	-	-	83 mg l^{-1}	60	-	-	-	-	-	5300	-	82- 94 mg l^{-1}	-	148 to 473 L $m^{-2}d^{-1}$	-	Almeida et al. (2017)
Filtralite® and gravel	tannery wastewater	horizontal	planted	-	-	-	-	-	-	-	-	-	-	*1800	*652	*3849	*1869	18, 8 and 6 cm d^{-1}	-	Calheiros et al. (2008)
LECA 10/20 mm	domestic wastewater	hybrid constructed wetlands	unplanted	36.1	63	22.9	77			1.2	89	11.8	78	19	91			7.4 $m^3 d^{-1}$ to 17.7 $m^3 d^{-1}$		Öövel et al. (2007)
LECA granules and powder	dairy industrial wastewater		unplanted						44.4		64.2	570	60	1220	68.4	2200	65.9		20 -120 h	Bahmanpour et al. (2017)

*kg ha^{-1} d^{-1}

Table 3. Removal efficiency for heavy metals and organic contaminants using LECA substrates.

Contaminant	% Removal efficiency	Comments	Reference
Pb Cd	93.7 89.7	Short contact time ranging from 1 to 2 hours for Pb and Cd adsorption. The removal rate of Cd and Pb gradually decreased with increase in contact time. Adsorption occurred at pH ranging from 3 to 10.	Malakootian et al. (2009)
Pb Cu	96 87	The presence of plants had no effect on Pb and Cu removal. Highest removal capacity observed for highly porous media.	Scholz and Xu (2002)
		<u>Organic contaminants</u>	
Oxytetracycline (antibiotic)	>97	Very high removal efficiency obtained in planted beds. Short contact time (within 3 days).	Dordio and Carvalho (2013a)
Polyphenols	80.3	A large proportion was removed after 3 days of contact time in planted beds.	
MCPA (herbicide)	77	High removal obtained in planted beds.	
Caffeine (wastewater indicator) Oxybenzone (sunscreen agent) and Triclosan (anti-bacterial agent)	19-85 61-97	High lipophilic compound removal is attributed to the presence of LECA.	Ferreira et al. (2017)
Polyaromatic hydrocarbons (PAHs): Phenanthrene Fluoranthene Pyrene	 92 93 94	Suggested LECA as alternative method for PAHs removal.	Nkansah et al. (2012)

Table 4 The chemical composition of LECA produced from clay, marine clay, and fabricator sludge.

Reference	Sharifnia et al., 2016	Kalhori et al., 2013	Laursen et al., 2006	
LECA raw material	100% clay	100% clay	90% marine clay+ 10% semiconductor production sludge	
			a	b
SiO_2	61.67	64.83	70.7	69.2
Al_2O_3	18.51	15.05	15.3	15.6
Fe_2O_3	6.14	7.45	4.5	4.42
MgO	3.97	3.67	1.02	1.03
CaO	3.5	2.98	3.8	3.97
K_2O	3.28	2.55	1.39	1.5
Na_2O	1.54	1.1	0.51	0.54
TiO_2	0.65	0.63	0.57	0.6
SO_3	0.23	0.11	1.5	2.22
P_2O_5	0.19	0.13	nd	0.026
SrO	0.13	-	0.026	0.023
Cl−	-	-	0.13	0.17
L.O.I	-	1.37	na	na
MnO	-	0.13	0.03	0.027
CuO	-	-	0.021	0.016
F	-	-	nd	0.21
ZnO	-	-	0.015	0.014
ZrO_2	-	-	0.101	0.053
BaO	-	-	0.36	0.31

Figures

Figure 1 Gravel-Steel Slag Horizontal Flow Constructed Wetland used as a polishing step at municipal wastewater treatment plant, Devizes, Wiltshire, UK. Effective operation of treatment cells (1) is ensured by flow meters (2) synchronized with separated distribution chambers (3). (Photo. F. Bydalek)

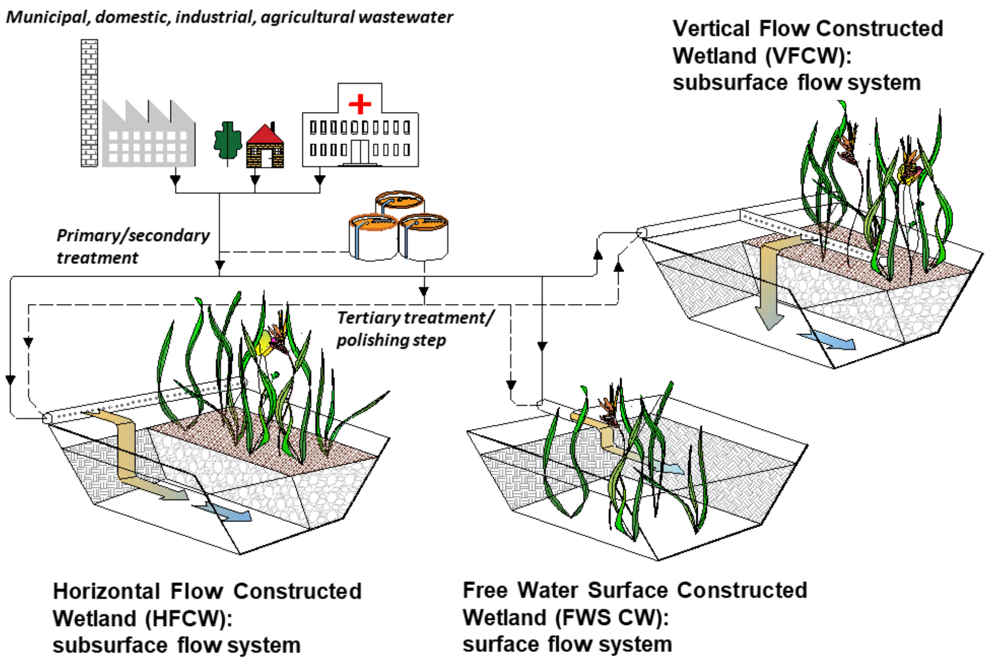

Figure 2 CWs are used for treatment of various types of wastewater, including rainwater, diluted municipal sewage and high strength industrial wastewater (i.e. effluents from slaughterhouses). CWs and can serve as either primary, secondary or polishing treatment step. There are three main types of constructed wetlands, classified based on the wastewater flow path: Free Water Surface CW (FWS CW), Horizontal Flow CW (HFCW) and Vertical Flow CW (VFCW).

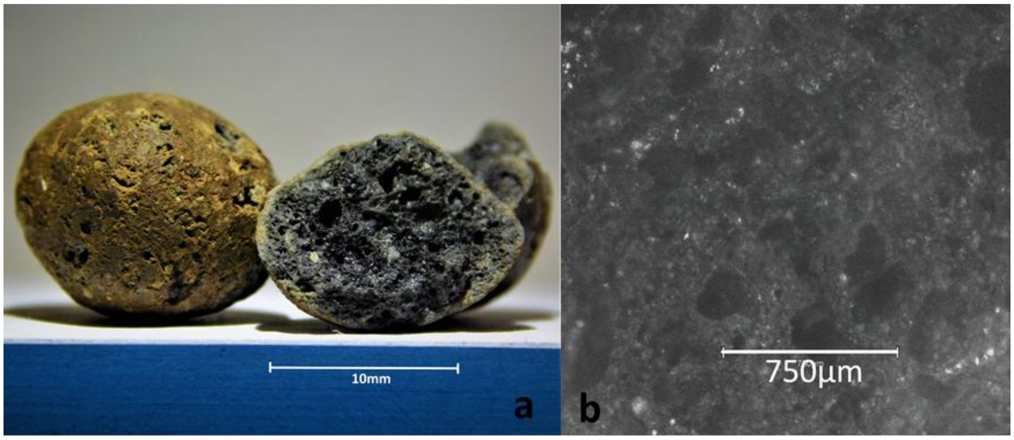

Figure 3 Macroscopic view (a) of intact and crushed LECA granule. Magnification of interior porous structures (b).

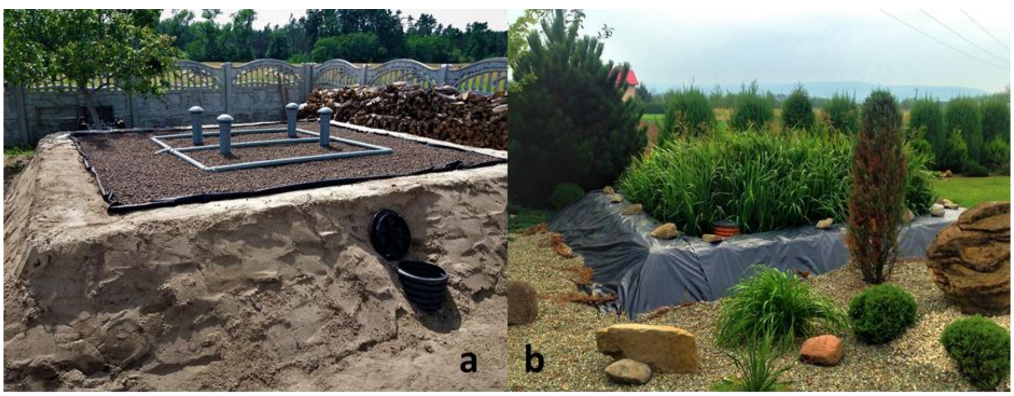

Figure 4 LECA-based Vertical Flow Constructed Wetland before (a) and after (b) commissioning. LECA has become one of the most commonly applied substrates for small scale, domestic CWs systems, which account for roughly 3000 units in Poland alone (*personal communication*). (Photo. F. Bydalek/ Ecoverde Engineering Office, Poland).

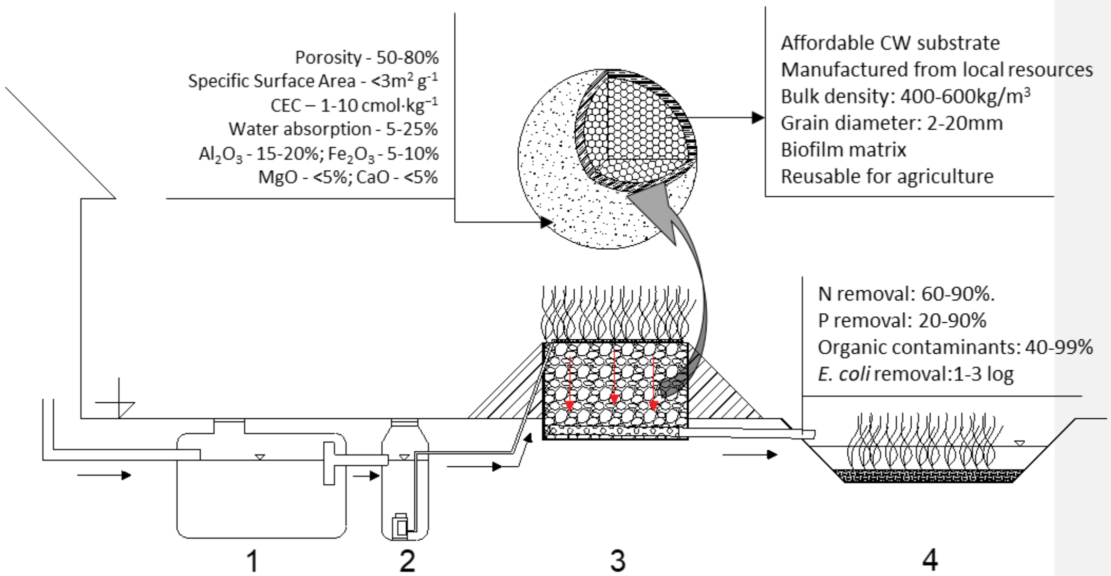

Figure 5 LECA properties enhance biological and physiochemical pollutant removal pathways in CWs. Schematic presentation of LECA-based VFCW designed for household use- 1) septic tank, 2) pump well, 3) elevated VFCW and 4) polishing pond.