



Full length article

Waste materials assessment for phosphorus adsorption toward sustainable application in circular economy

Magda Kasprzyk*, Krzysztof Czerwionka, Magdalena Gajewska

Gdansk University of Technology, Faculty of Civil and Environmental Engineering, Department of Water and Wastewater Technology, Narutowicza st. 11/12, 80-233 Gdansk, Poland

ARTICLE INFO

Keywords

Circular economy
Material recycling
Phosphorus removal
Sorption kinetic
Wastewater treatment
Zero waste

ABSTRACT

Phosphorus is the main determinant of nutrient enrichment in the water bodies. Many resources including nutrients may be shortly exhausted, assuming current consumption. This scenario leads to growing interest in resources recovery and/or reuse, which together with sustainable energy consumption and waste reduction are the main courses of the circular economy. Usage of coagulants in wastewater treatment plants (WWTP) does not allow phosphorus to be recovered from the sludge. An alternative method with recovery potential is the application of adsorbents. Many adsorbents have been approved for P-removal, however some of them are expensive. Response to that issue may be the application of waste materials and byproducts often intended for disposal/utilization. The main objective was to assess the P-removal efficiency of the waste materials/byproducts for application in small WWTP and eco-technologies like treatment wetlands as replacement of commercial adsorbents and alternative for conventional (chemical) treatment for potential phosphorus recovery. The materials investigated were (1) an expensive man-made product, (2) a byproduct with a high content of calcium oxide, and (3) drinking water treatment residuals (DWTRs) as beds from filters used to remove iron. Research was conducted in non-flow condition with various initial concentrations. The highest efficiency was received for fine-grained byproduct (9.58 mg/g), for commercial adsorbent LMB - 9.11 mg/g. The DWTRs adsorption was oscillated around 4.5 mg/g. These results confirmed the possibility of applying waste materials and byproducts for P-removal according to the circular economy idea. Further research should include implications of waste materials application to determine P-recovery concept.

1. Introduction

The circular economy concept is developed from increasing the consumption of limited natural resources. Unsustainable resource exploitation results in environmental degradation, imbalance of biodiversity, and water bodies pollution (Acquaye et al., 2018; Caniato et al., 2012; Moktadir et al., 2020a). That leads to growing interest in recovery and/or reuse of resources, which together with sustainable energy consumption and waste reduction are the main courses of the circular economy (Zamparas et al., 2020). The Circular Economy idea obligates to maintain resources and materials in the environment as long as possible and turn waste into a product for another use. The possibility of potential element recovery is attracting much more attention due to the European Commission action plan for the concept of a Circular Economy (COM/2015/614 final).

1.1. Phosphorus removal and recovery – Circular economy challenges

Phosphorus compounds are considered to be the main determinant of nutrient enrichment in the water bodies, which are the main cause of the increasing growth of algae and cyanobacteria (Asfar and Groves, 2009; Smith et al., 1999; Van Oosterhout and Lüriling, 2013; Van Oosterhout et al., 2014). The presence of phosphorus in surface water, despite being a natural one, is mostly caused by the over-fertilisation of agricultural areas, but also comes from untreated or inadequately treated domestic and industrial wastewater discharged directly to the aquatic ecosystems (Zamparas et al., 2012).

The conventional phosphorus removal in the wastewater treatment plants (WWTP) is frequently provided by chemical reagents such as ferric sulphate and chloride or aluminium sulphate. Such methods might be effective, though they are not economically justified in smaller towns (up to 10 000 person equivalent) and rural areas, where treatment wetlands are more suitable (Kadlec and Wallace, 2009; Vy-

* Corresponding author.

E-mail addresses: magkaspr@pg.edu.pl (M. Kasprzyk); kczer@pg.edu.pl (K. Czerwionka); mgaj@pg.edu.pl (M. Gajewska)

mazal, 2011). Despite the phosphorus removal issue, many resources including nutrients are increasingly exploited and may be shortly exhausted, assuming current consumption of raw materials (Guerra-Rodriguez et al., 2020). Increasing awareness of the limited resources of phosphorus brings the necessity to discover a sustainable way for P-removal from the wastewater towards its reuse for industry or agriculture. It is worth pointing out, that applying coagulants does not allow the recovery of phosphorus from the sludge, so it is not bioavailable for plant growth (Guerra-Rodriguez et al., 2020).

The circular economy requirements have initiated research in P-recovery. To date, many mechanisms of phosphorus recovery have been discovered (Guerra-Rodriguez et al., 2020; Hukari et al., 2016; Leyva-Diaz et al., 2020; Sfez et al., 2019). Main forms of recovery technology may include chemical precipitation, wet-chemical process, thermochemical process and adsorption, both from the liquid and sludge phase (Amann et al., 2018; Cornel and Schaum, 2009; Egle et al., 2016). The efficiency of such methods is very high and may reach over 90% (Egle et al., 2015; Ye et al., 2017).

A promising alternative method in terms of recovery potential is the application of sorption materials. Many adsorbents of different origins, both natural and synthetic, have been approved for the ability to phosphorus removal (Cucarella and Renman, 2009; Douglas et al., 2016; Johansson Westholm, 2006; Vohla et al., 2011). Unfortunately, some of them are expensive or locally inaccessible that their application is simply not economically worthwhile. A response to that issue but also innovative approach may be the application of byproducts or waste materials, which also follow the assumption of a circular economy.

Many recent investigations are focused on waste materials usage for wastewater treatment including natural and inorganic sources, municipal waste, agricultural and industrial byproducts and waste (Hossain et al., 2020), as well as the mining tailings from the iron mine for industrial effluent treatment (adsorption of metals like chromium, copper, zinc) (Izidoro et al., 2019). Moreover, the mining waste recovery potential is also investigated (Perez et al., 2020). Some of researchers concentrate on finding waste material/industrial byproduct strictly for phosphorus removal as fly ash, blast furnace slag or oil shale (Drizo et al., 1999, 2006; Vohla et al., 2011).

1.2. Potential waste materials for P-removal

Drinking water treatment residuals (DWTRs) are an unavoidable byproduct generated during the drinking water treatment process and are often stored in landfills or as waste. However, DWTR disposal costs may increase due to the increased production of those byproducts (Ahmad et al., 2016). Recently, the reuse of residuals, instead of utilisation, becomes an alternative and interesting method. Aluminium and iron salts, used for potable water treatment, result in enhanced porosity and adsorption capacity (Ippolito et al., 2011). Furthermore, thus far DWTRs were successfully applied for the removal of P (Oliver et al., 2011, Wang et al., 2011), arsenic (Nagar et al., 2010), lead (Zhou and Haynes, 2011), chromium (Zhou and Haynes, 2011), copper (Lin et al., 2014), nickel (Chiang et al., 2012), zinc (Chiang et al., 2012), organic substances (Hu et al., 2011) and others.

Drinking water treatment residuals (DWTRs) may be an effective solution due to their high P-sorption capacity and non-hazardous character. Moreover, they can be divided into (i) a sludge obtained after coagulation, thus DWTRs are very often rich in Fe or Al oxides due to the Fe and Al coagulant application during the drinking water treatment process (Dayton and Basta, 2005). Moreover, DWTRs can be integrated with agricultural soils (Callahan et al., 2002; Dayton and Basta, 2005; Wang et al., 2013), applied to edge of a stream to reduce phosphorus loss and preserve adjacent waters (Dayton and Basta, 2005; Penn and Bryant, 2006) or applied to impacted cattle loafing areas prior to rainfall, which can mitigate considerable P amounts in runoff (Penn and Bryant, 2006). Research performed by

Wang et al. (2018), in a one-year continuous flow column test, indicates that the application of DWTRs significantly increased P removal effectiveness and reduced algae growth (*Microcystis aeruginosa*).

Furthermore, DWTRs are suitable for application as constructed wetland substrates to facilitate phosphorus removal from wastewater (Babatunde et al., 2009; Bai et al., 2014). Research on additional rechargeable cells, filled with sorption material for phosphorus removal, have been carried out (Arias et al., 2003; Zhu et al., 2003). DWTRs, as the contents of such cells, might be an inexpensive, sustainable method for improving non-waste technology like treatment wetlands for water pollution control.

Another kind of DWTR is (ii) a waste material obtained from the exchange of the deferrisation filter filling in underground water treatment plants, which is usually motivated by technological requirements. There is no available research on this particular sort of material, although a study on a steel industry byproduct used for phosphorus removal from wastewater revealed that the mechanism of P-removal is dominated by adsorption onto ferric oxyhydroxides and associated with amorphous Fe phases (Pratt and Shilton, 2010).

The goals of this study were to:

- 1) compare a commercial sorbent, with known and confirmed effectiveness in P-removal with materials often disposed of as wastes via landfill,
- 2) establish the possibility of waste material/byproduct usage as replacement of commercial adsorbents for P-removal from wastewater according to circular economy requirements.

The chosen materials were (1) a lanthanum modified bentonite clay (LMB), an expensive, man-made product, (2) a byproduct remaining after thermal treatment of carbonate-siliceous rock with a high content of calcium oxide (CaO), and (3) drinking water treatment residuals (DWTRs) as beds from filters used to remove iron from underground water at various drinking water treatment plants. Study goals were accomplished by:

- 1) quantifying the P-sorption capacity of LMB, byproduct and DWTRs
- 2) fitting experimental data with adsorption isotherms (Langmuir, Freundlich, BET and Temkin)
- 3) obtaining kinetic parameters from pseudo-first order and pseudo-second order models.

2. Materials and methods

Sorption materials of different origin and composition were investigated. Several laboratory trials were carried out to evaluate the possibility of phosphorus removal by the usage of selected sorption materials. Graphics (Fig. 1) present research procedure.

2.1. Materials

(1) lanthanum-modified bentonite clay (LMB) is an adsorbent developed by the Land and Water Division of Australia's CSIRO (Commonwealth Scientific and Industrial Research Organization) (Douglas et al., 1999). The aim of the application of the material is to manage the massive loading of phosphorus in aquatic systems. LMB is characterised by phosphorus binding in the molar ratio of P:La equal to 1:1, therefore during the reaction, there is no byproduct formation. Moreover, rhabdophane ($\text{LaPO}_4 \cdot n\text{H}_2\text{O}$), which is formed in the binding reaction, is a stable, resistant rare earth mineral (Douglas et al., 2016). The LMB consist mostly SiO_2 (63.36%) and Al_2O_3 (14.73%) (Haghseresht et al., 2006, 2009). Characteristic of the material is presented in Table 1.

Powder Rockfos® byproduct (2) obtained after the decarbonisation process (firing at high temperature) of natural silica rock-carbonate was considered as a waste material towards circular economy regula-

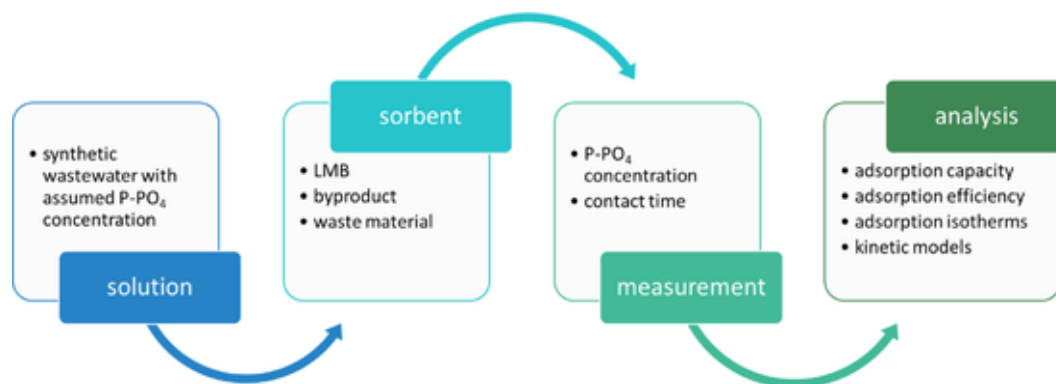


Fig. 1. Graphical stepwise procedure of materials and method.

Table 1
Main chemical compounds and physical properties of tested materials.

	(1) LMB	(2) Rockfos byproduct	(3a) DWTRsReda	(3b) DWTRsLetniki	(3c) DWTRsZaspa Wodna
main chemical compounds					
SiO ₂	~ 63%	–	–	–	–
CaO	–	~ 80%	–	–	–
La ₂ O ₃	0.058%	–	–	–	–
Fe content (mg Fe/g _{d.w.})	–	–	27.49	98.97	42.51
Mn content (mg Mn/g _{d.w.})	–	–	1.85	50.27	5.37
physical properties					
Average particle size (mm)	2 – 4 × 1–3	0–2	2.08	1.21	1.30
pH	6.8–7.5	11– 12	–	–	–
Total pore volume (cm ³ /g)/ Porosity (%)	0.171	~ 50%	47.8	61.2	57.0
Appearance	brown free-flowing granules	white fine-grained particles	quartz sand grains		

tions. Rockfos® is a relatively novel material produced by Kufel Ceramics Company in Poland, and is highly reactive to the phosphorus due to a substantial amount of calcium and silicon. Commercial Rockfos® particle size is 2–5 mm and can be used to build filters for phosphorus and bacteriological contaminant removal from domestic and industrial wastewater. During the phosphorus removal process, it is bound by sorption to calcium phosphates Ca₃(PO₄)₂ (Rockfos®, 2020). The analysed byproduct is characterised by a very fine average particle size of 0–2 mm, with a porosity of over 50%, and significant content of calcium oxide, close to 80% (Table 1). Due to the composition of the waste material, the pH is alkaline and equal to 11–12.

To pursue a “no waste” policy, drinking water treatment residuals as a third material (3) (DWTRs) from three different underground water treatment plants were considered during the following investigation. Specific parameters of DWTRs are shown in Table 1.

(3a) DWTRs from “Reda” DWTP, where the treated water is characterised by an elevated content of iron (up to 2.6 mg Fe/L) and manganese (up to 0.3 mg Mn/L). Material from deironing pressure filters was stored in the open air for 2–3 years. The filtration material consisted of quartz sand fraction (1.4–2.1 mm) and gravel fraction (4.0–7.0 mm). To use the DWTRs in the laboratory study, they were air-dried at a temperature 20–23 °C, sieved through a sieve with a 4.0 mm mesh size and stored in open vessels. The iron oxide and manganese oxide layer on the quartz sand grains was wear and shatter-resistant. The “Reda” DWTRs are characterised by the lowest content of iron and manganese.

Water supplied to the “Letniki” DWTP exceeds the acceptable concentration level of iron (3.2–8.1 mg Fe/L), manganese (0.5–2.28 mg

Mn/L) and ammonia nitrogen (1.2–2.88 mg N—NH₄/L). (3b) The “Letniki” DWTRs were collected after over a year of working as filter filling. To use the DWTRs in the laboratory study, they were dried out at a temperature of 20–23 °C and stored in open vessels. The filter medium was composed of quartz sand fraction (0.8–1.9 mm). The content of iron and manganese was the highest from the analysed DWTRs. The layers on the quartz sand grains were easily shattered and form a fine powder of the iron and manganese oxides.

The DWTP “Zaspa Wodna” treats water with elevated concentration of iron (0.7–1.5 mg Fe/L) and manganese (0.15–0.26 mg Mn/L). (3c) The “Zaspa Wodna” DWTRs consisted of quartz sand fraction (0.8–1.4 mm). To use the DWTRs in the laboratory study, they were dried out at a temperature of 20–23 °C and stored in open vessels. The unique feature of the “Zaspa Wodna” DWTRs was a high concentration of iron while maintaining a relatively low concentration of manganese.

2.2. Methods

The investigation was divided into two separate types of research. The first study was carried out to determine the specific parameters of the adsorption isotherms (Langmuir, Freundlich, BET and Temkin). The second study was performed to obtain kinetic parameters from the pseudo-first (PFO) and pseudo-second order kinetic model (PSO). The selection of sorption kinetic models was motivated by comparison simplicity as those methods are the most often used by researchers (Dayton and Basta, 2005; Del Bubba et al., 2003; Ho and McKay, 1998; Leader et al., 2008; Li et al., 2018; Liu and Zhang, 2015; Zamparas et al., 2020). The scope of analysis is shown in Table 2.

Table 2
The experiment set up of conducted studies.

adsorbent	solution	Temperature	study	volume	dose	time	PO ₄ -P
LMB / powder Rockfos® byproduct	KH ₂ PO ₄	20 °C	adsorption isotherms	1.0 L	10.0 g	1 h	5–100 mg/L
			kinetic model	1.0 L	1.0 g	5 h	10 mg/L
DWTRs	NaH ₂ PO ₄	25 °C	adsorption isotherms	0.15 L	15.0 g - 0.4 g	24 h	5–300 mg/L
			kinetic model	0.15 L	15.0 g	24 h	15 mg/L

Each stage was performed with a batch reactor model in static conditions. Prior to the addition of the adsorbent, it was dried at a temperature of 105 °C. Following a contact time period, samples were filtered through a 0.4 µm Millipore filter and analysed for PO₄-P concentration using a HACH Lange DR 3900 laboratory VIS spectrophotometer with RFID and cuvette tests in case of adsorbents (1) and (2). Samples with DWTRs were measured using an ammonium molybdate spectrometric method (wavelength – 690 nm).

The adsorption capacity was determined according to the following Eq. (1) (Huang et al., 2015; Nastawny et al., 2015; Liu and Zhang, 2015; Zamparas et al., 2012):

$$q = \frac{(C_0 - C) \cdot V}{m} \quad (1)$$

where: q – adsorption capacity [mg/g], V – volume of the solution [L], C₀ – initial concentration of PO₄-P [mg/L]; C – equilibrium concentration of PO₄-P [mg/L], m – the mass of adsorption material [g].

2.3. Adsorption model

P-removal parameters and maximum adsorption capacity at equilibrium were determined by the correlation between the amount of adsorbed phosphates q [mg/g] and the final concentration of phosphates C [mg/L]. The Langmuir model assumes monolayer adsorption; the surface of adsorption is homogenous with no adsorbed particle interactions. The Freundlich model assumes multilayer adsorption on a heterogeneous surface (Li et al., 2018; Song et al., 2011; Wang et al., 2016; Xu et al., 2013; Zamparas et al., 2012). The adsorption isotherms are described by the following mathematical equations (Del Bubba et al., 2003; Liu and Zhang, 2015; Xu et al., 2013; Zamparas et al., 2012):

Langmuir isotherm

$$q = \frac{b \cdot q_m \cdot C}{1 + b \cdot C} \quad (2)$$

where: q – sorption capacity [mg/g]; b – constant in Langmuir model of adsorption; C – concentration of PO₄³⁻-P at equilibrium [mg/L], q_m – maximum sorption capacity [mg/g].

In linear form for materials (1) and (2):

$$\frac{1}{q} = \frac{1}{q_m \cdot b} \cdot \frac{1}{C} + \frac{1}{q_m} \quad (3a)$$

In linear form for materials (3a,b,c):

$$\frac{C}{q} = \frac{1}{q_m \cdot b} + \frac{C}{q_m} \quad (3b)$$

Freundlich isotherm

$$q = k \cdot C^{1/n} \quad (4)$$

where: q – sorption capacity [mg/g]; C – concentration of PO₄³⁻-P at equilibrium [mg/L]; k, 1/n – constants in Freundlich model of adsorption.

In linear form:

$$\log q = \log k + (1/n) \log C \quad (5)$$

Brunauer, Emmet, and Teller (BET) established the multilayer model of adsorption, where the surface is divided into some empty sites and some covered by molecule layers (Tan and Haver, 2004). Similarly to the Langmuir isotherm, the BET adsorption model assumed no interactions between molecules (Gobin, 2006). The BET model can be written as (Ebadi and Soltan, 2009; Gobin, 2006):

BET isotherm

$$q = \frac{B \cdot q_m \cdot \frac{C}{C_n}}{\left(1 - \frac{C}{C_n}\right) \cdot \left[1 + (B - 1) \cdot \frac{C}{C_n}\right]} \quad (6)$$

In linear form:

$$\frac{\frac{C}{C_n}}{q \left(1 - \frac{C}{C_n}\right)} = \frac{1}{B \cdot q_m} + \frac{B - 1}{B \cdot q_m} \cdot \frac{C}{C_n} \quad (7)$$

where: q – sorption capacity [mg/g]; C – concentration of PO₄³⁻-P at equilibrium [mg/L]; q_m – maximum sorption capacity [mg/g]; B – constant in BET model of adsorption; C_n* – saturation concentration of the solution [mg/L], according to (Buhl et al., 1994):

* for KH₂PO₄ in 20 °C – C_n = 51,481 mg/L for NaH₂PO₄ in 25 °C – C_n = 247,610 mg/L

Temkin isotherm allows to evaluate the influence of temperature in adsorption process and assumes the linear decrease in the heat of adsorption in the opposite to Freundlich isotherm (logarithmic fall) (Han et al., 2005; Izidoro et al., al.,2019; Xu et al., 2013). The interaction between the adsorbent and the adsorbate is take into consideration in Temkin model assumption. Its suitable for intermediate concentrations of adsorbate due to reject high and low concentration values (Al-Ghouti and Da'ana, 2020; Ayawei et al., 2017; Febrianto et al., 2009; Prasanna Kumar et al., 2006; Singh et al., 2018). Temkin model is the following:

Temkin isotherm

$$q = \frac{RT}{b_T} \ln A_T C \quad (8)$$

In linear form:

$$q = \frac{RT}{b_T} \ln A_T + \frac{RT}{b_T} \ln C \quad (9)$$

where: q – sorption capacity [mg/g]; C – concentration of $\text{PO}_4^{3-}\text{-P}$ at equilibrium [mg/L]; R – universal gas constant [8.314 J/mol K], T – temperature [K]; b_T – Temkin constant related to the heat of adsorption [kJ/mol], A_T – Temkin isotherm equilibrium binding constant [L/g].

2.4. Kinetic model

To develop rate constant parameters of the adsorption mechanisms, pseudo-first and pseudo-second order model were determined. According to Lagergren, pseudo-first order model describe process of liquid–solid phase adsorption (Berkessa et al., 2019; Febrianto et al., 2009; Ho and McKay, 1999).

$$\frac{dq_t}{dt} = k_1 (q_e - q_t) \quad (10)$$

where: q_t – adsorption capacity of material at time t [mg/g], q_e – adsorption capacity of material at equilibrium [mg/g], k_1 – equilibrium rate constant of pseudo-first order reaction [1/min].

For boundary condition as $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_e$:

$$\log \left(\frac{q_e}{q_e - q_t} \right) = \frac{k_1}{2.303} t \quad (11)$$

Rate constants were estimated due to model transformation to linear form and linear plot $\log(q_e - q_t)$ against t . Eq. (11) in linear form is:

$$\log (q_e - q_t) = \log q_e - \left(\frac{k_1}{2.303} \right) t \quad (12)$$

The kinetic model constant parameters (k_1 , q_e), the determination coefficients R^2 and the correlation coefficients r were acquired.

The phosphate adsorption mechanism may be described also by the pseudo-second order model. This particular kinetic mechanism was established to determine the rate constant of adsorption and the equilibrium adsorption capacity. A rate parameter, k , has been defined and used to describe the sorption of acidic and basic dyes (Ho and McKay, 1998). The pseudo-second order model assumed that the adsorption follows second order chemisorptions (Ho and McKay, 1999). Moreover, it is used to define the chemisorption of pollutants from aqueous solution onto the adsorbent (Li et al., 2015). The pseudo-second order model describing the adsorption process is the following (Ho and Chiang, 2001; Haghseresht et al., 2009; Liu and Zhang, 2015; Ross et al., 2008):

$$\frac{dq_t}{dt} = k_2 (q_e - q_t)^2 \quad (13)$$

where: q_t – adsorption capacity of material at time t [mg/g], q_e – adsorption capacity of material at equilibrium [mg/g], k_2 – equilibrium rate constant of pseudo-second order reaction [g/mg·min].

With the initial and boundary condition as $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_e$, Eq. (13) convert into:

$$\frac{1}{q_e - q_t} = \frac{1}{q_e} + k_2 t \quad (14)$$

Formula (14) transformation to a linear model is shown as:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (15)$$

From developing a linear function t/q_t against time, the kinetic model constant parameters (k_2 , q_e), the determination coefficients R^2 and the correlation coefficients r were obtained.

3. Results and discussion

The maximum adsorption capacity obtained during conducted trials for each of the analysed material is presented in Table 3. In any case, the maximum adsorption capacity of phosphates was achieved for the highest considered concentration of $\text{PO}_4\text{-P}$: ~100 mg/L for LMB and Rockfos® byproduct, ~300 mg/L for DWTRs). As it is shown in Table 4, the maximum P-capacity was received for fine-grained Rockfos® byproduct and equal to 9.58 mg/g. Similarly, a high result was noted for commercial adsorbent LMB (9.11 mg/g). The research described by Kasprzyk and Gajewska (2019), carried out in multiple uses of those materials, has shown significant ability to remove phosphates. The adsorption capacity achieved for LMB was equal to 5.6 mg/g, while for the Rockfos® byproduct reached over 45.6 mg/g. According to Haghseresht et al., 2009, P-sorption of LMB cannot exceed 10.6 mg/g due to its chemical composition (approx. 49 mg La/g of LMB and 1:1 molar ratio of La and P). In the case of (2) Rockfos® byproduct with a high content of CaO, there is no study developing adsorption ability of this particular material, although other Ca-rich natural sorbents as Opoka and Filtra P, were investigated (Brogowski and Renman, 2004; Bus and Karczmarczyk, 2014; Cucarella et al., 2007). Brogowski and Renman (2004) discovered that the heating process of Opoka results in a significant increase of $\text{PO}_4\text{-P}$ sorption capacity up to 119.6 mg/g for a heating temperature of 1000 °C. Moreover, substantial P-adsorption capacity of this material can be related to very fine granulation (powder fraction), which gives a large specific surface area for phosphate binding.

The best of DWTRs was “Zaspa Wodna”, with an adsorption capacity at the level of 4.88 mg/g, the lowest – “Letniki” (4.24 mg/g), although each of the DWTRs obtained average but satisfactory results compared with DWTRs tested in other research. According to Cucarella and Renman (2009), P-sorption capacity obtained for Fe-coated sand and brick may be 1–10 mg/g, while Fe-DWTRs achieved a result at the level of 0.95 mg/g (Leader et al., 2005, 2008). Furthermore, a recent study performed by Li et al. (2019), presented promising results for powder fraction and a granular fraction of DWTR where P-adsorption capacity reaches 23.92 mg/g and 21.56 mg/g, respectively for the highest phosphate concentration equal to 1000 mg/L. Column tests with glass beads filled with DWTR ensured an output with P-concentration lower than 0.01 mg/L (Wang et al., 2018).

As an advantage, pH level can be considered, both adsorbent pH as well as the operative pH range of the solution. The pH of the LMB is neutral (6.8–7.5). Moreover, no significant influence of LMB on solution pH was discovered (Kasprzyk et al., 2018). In regard to effective phosphate removal, maximum efficiency is achieved within a pH of 5–7, but a sufficient reduction level is obtained between a pH level of 5–9

Table 3
Maximum achieved adsorption capacity of each tested material.

	initial $\text{PO}_4\text{-P}$ mg/L	volumel	timeh	doseg	adsorption capacitymg/g	efficiency%
(1)	95.59	1.00	1	10.0	9.11	95.3
(2)	97.33	1.00	1	10.0	9.58	97.8
(3a)	294.0	0.15	24	0.4	4.58/0.06*	4.1/92.8*
(3b)	302.0	0.15	24	0.4	4.24/0.06*	3.7/94.9*
(3c)	298.0	0.15	24	0.4	4.88/0.06*	4.4/91.9*

* adsorption capacity and maximum efficiency obtained for initial $\text{PO}_4\text{-P}$ 5 mg/L and 15 g adsorbent dose.

Table 4
Parameters of the linear Langmuir, Freundlich, BET and Temkin isotherms.

	Langmuir isotherm				Freundlich isotherm			
	b	q _m	R ²	r	k	1/n	R ²	r
(1)	0.0944	158.73	0.81	0.89	5.48	0.51	0.73	0.92
(2)	0.0084	256.41	0.99	0.97	2.84	1.34	0.97	0.99
(3a)	0.0058	6.57	0.74	0.99	0.074	0.71	0.99	0.99
(3b)	0.0050	6.36	0.70	0.99	0.071	0.69	0.97	1.00
(3c)	0.0052	6.97	0.72	0.98	0.068	0.73	0.99	1.00
	BET isotherm				Temkin isotherm			
	q _m	B	R ²	r	b _T	A _T	R ²	r
(1)	10.55	73,220.2	0.91	0.94	1.40	46.86	0.95	0.98
(2)	11.11	64,547.6	0.89	0.93	0.67	3.29	0.74	0.87
(3a)	6.56	1444.2	0.74	0.99	3.64	0.43	0.76	0.87
(3b)	6.35	1242.3	0.70	0.99	4.12	0.45	0.72	0.85
(3c)	6.96	1293.6	0.72	0.98	3.49	0.39	0.75	0.87

(Copetti et al., 2015; Ross et al., 2008). Also in several studies, no negative and harmful effects on aquatic organisms were discovered (Grove, 2010; van Oosterhout et al., 2014). However, LMB is a commercial product (Phoslock®), which is linked to investment and maintenance costs (Kasprzyk et al., 2018).

It is worth noticing that the most effective material was a waste product, which was often utilised before. Therefore the application of such material is not only efficient but also follows the circular economy idea. The disadvantage of the Rockfos® byproduct is the high pH (11–12), which causes an alkaline solution. Moreover, multiple uses of the adsorbent slightly decreased the solution pH, but it was still close to 9 (Kasprzyk and Gajewska, 2019). Similar effects were described for heated Opoka, where the solution pH reached 12.6 (Brogowski and Renman, 2004).

3.1. Isotherms of adsorption

Data obtained was fitted by the adsorption isotherm of Langmuir, Freundlich, BET and Temkin. The linear form of the Langmuir isotherm fitted the achieved sampling points for all analysed materials. The determination coefficient was about 0.7 for DWTRs, 0.8 for LMB, and the highest value was obtained for the Rockfos® byproduct, equal to 1.0. The correlation coefficient shows relatively good matching to the approximated data.

Specific parameters of the adsorption isotherms are presented in Table 4. The maximum adsorption capacity achieved from the Langmuir isotherm was the highest for the Rockfos® byproduct and equal to 256.4 mg/g. As a confirmation of the significant ability of P-removal by Ca-rich materials, a study from Brogowski and Renman (2004) can be referred to, as they obtained 120 mg/g adsorption capacity for Opoka heated in 900 °C. LMB product theoretical q_m reached 158.7 mg/g, while research performed by Haghseresht et al. (2009) shows that maximum sorption capacity of LMB ranged from 9.5 to

10.5 mg/g. Zamparas et al. (2012) revealed q_m value for LMB around 11.6 mg/g. For DWTRs, that parameter was relatively low, not exceeding 7.0 mg/g, whilst from studies conducted by Li et al. (2018 and 2019), the maximum adsorption capacity obtained from the Langmuir isotherm was equal to 26.5 mg/g for powder DWTRs and 19.70–23.3 mg/g for granular DWTRs.

The Freundlich isotherm of adsorption shows the best matching of a linear function to the data obtained. For all tested materials, the determination coefficient amounted to 1.0, except LMB (R²=0.73), which may indicate that the Freundlich isotherm is more suitable for adsorption character description. The correlation coefficient in each case exceeds 0.9 and confirms the good approximation of presented results with P-capacity calculated from the Freundlich equation (Table 4).

To verify the maximum theoretical sorption capacity, a transformed linear BET equation was also used. The results of the calculated q_m are shown in Table 4. For DWTRs, that parameter was almost equal to the maximum sorption capacity achieved for the Langmuir isotherm, while for LMB and Rockfos® byproduct, the P-capacity was significantly lower, 10.55 and 11.11 mg/g, respectively. The BET function matches the measured points relatively well, due to the obtained determination coefficients values for the LMB and byproduct R²=0.9, and DWTRs around 0.7. The correlation coefficient in each case was above 0.9.

From the Temkin isotherm, rate constant parameters b_T, A_T together with determination and correlation coefficients were evaluated. amongst investigated models, Temkin model is in average agreement with the experimental data with R² equal to 0.7 (only for LMB R²=0.98). In the case of the correlation coefficient, achieved results were worse than those for other isotherms. Reported bonding energy (b_T) range for ion-exchange is in the range of 8–16 kJ/mol, for physisorption processes – less than 40 kJ/mol. Values obtained in the present study (0.67 – 4.12 kJ/mol) can indicate that process involve chemisorption and physisorption (Liu and Zhang, 2015). Also, the consistent distribution of energy binding is an assumption of the Temkin isotherm (Izidoro et al., 2019).

3.2. Adsorption kinetics of phosphates

The pseudo-first order kinetic model fits relatively well acquired experimental data with R² over 0.9 for almost all tested materials. The correlation coefficient indicates strong negative correlation – r close to –1.0. The equilibrium capacity of adsorption obtained from pseudo-first order model was the highest for byproduct (7.9 mg/g). The all results of the kinetic parameters are given in Table 5.

The pseudo-second order kinetic model presents good compliance with the obtained results for each of the investigated materials. The determination coefficients for the linear function were very high and close to 1.0 for all adsorbents in these study.

The sorption capacity at equilibrium was the lowest from all the others obtained in this study. For the LMB and Rockfos® byproduct, equilibrium adsorption capacity of phosphates was equal to 4.3 mg/g and 4.5 mg/g respectively. DWTRs were shown to have a significantly lower P-capacity from the BET equation, at the level of 0.1 mg/g (Table 5).

Table 5
Kinetic parameters for the phosphates adsorption.

	pseudo-first order model				pseudo-second order model			
	q _e mg/g	k ₁ min ⁻¹	R ²	r	q _e mg/g	k ₂ g/mg min ⁻¹	R ²	r
(1)	6.40	0.0010	0.86	–0.93	4.31	0.019	1.00	1.00
(2)	7.90	0.0013	0.99	–0.99	4.48	0.007	0.97	0.98
(3 a)	0.17	0.0048	0.96	–0.98	0.10	0.062	0.98	0.99
(3 b)	0.17	0.0049	0.96	–0.99	0.11	0.040	0.97	0.98
(3 c)	0.18	0.0049	0.96	–0.99	0.13	0.040	0.97	0.98

3.3. Cost analysis and possible benefits

As predicted, the commercial adsorbent (LMB) displayed a substantial ability to remove phosphorus compounds and, because of this, was found to be successful in the reversal of eutrophication, and has been used to reduce algae growth in about 200 water bodies all over the world (Copetti et al., 2015; Douglas et al., 1999, 2016). According to Kasprzyk et al., 2018, the annual maintenance of the P-removal additional unit in a small treatment wetland can reach 14,000€, 1 kg of removed PO₄-P may cost 1400€, while 1m³ of treated effluent can cost 6€.

Furthermore, the production of Rockfos® results in the production of 30% of byproduct, which means 300 kg of possible adsorption material is collected from each produced ton of Rockfos®. Considering the adsorption capacity of the byproduct achieved during this investigation, a ton of Rockfos® could give the possibility to remove almost 2.9 kg of phosphates free of charge. Moreover, the commercialisation of such a material could bring the opportunity of additional benefits for the producer. However, preparation for sale is connected with specific actions to maximise the byproduct collection and keep it as clean as possible during the production process and to ensure stable storage conditions.

DWTRs, obtained from an exchange of filling from filters for iron removal, are not regularly produced waste material, due to the fact that the exchange of the filter bed is usually caused by technological issues. DWTRs do not have specific regulations for the disposal or storage of such materials, thus it would be required to establish exchange/storage procedures for a possible sales opportunity. Such beds are created during the typical operation of water iron removal filters, and in the event of a market emergence, it would be possible to produce DWTRs in an underground water treatment plant.

The presented investigation revealed the hypothetical possibility of the replacement of an expensive adsorbent with waste material or a byproduct. It is difficult to assess the cost of using such material, due to the fact that they do not exist in the trade market. It can be assumed that the application of a waste or byproduct is free of charge and it can even reduce disposal costs for the producer. Moreover, studies that confirm the removal efficiency of materials designated for utilisation, may reverse that policy and create a sustainable treatment method and minimise generation of waste.

3.4. Circular economy – Limitation of use

Table 6 presents predicted limitation of use for analysed materials. Factors as price, efficiency in P-removal, pH, availability (local access, transportation) and possible P-recover/reuse for circular economy compliance have been taken into account. The most favourable seems to be byproduct of Rockfos® material due to fulfilling all assumed conditions. *LMB does not fit CE since P-removal process results in highly insoluble compound (rhabdophane). **Rockfos® byproduct might be potential source of phosphorus, calcium phosphate is available for plants, although high pH may be limitation and obligates to use it for acidic soil. However, there is not much research about this particular material to confirm recover/reuse possibility. ***DWTRs can enrich soil with ferric, manganese and phosphorus, nevertheless P-removal efficiency is

Table 6
Limitation of use for tested materials.

	price	efficiency	pH	availability	CE(recover/reuse)	Rating
(1) LMB	-	+	+	+	.*	3/5
(2) byproduct	+	+	+/-	+	+ **	5/5
(3) DWTRs	+	-	+	-	+ ***	3/5

*, **, *** explanation placed in the text.

significantly lower than others tested materials and its bioavailability is difficult to predict.

To pursue the concept of the circular economy, the waste products used for the filling of additional P-removal filters can be exploited as P-rich material for agriculture purpose, after exhaustion of material sorption capacity. However, such action is subject to further studies to investigate and exclude potential hazardous risks. DWTRs can be considered to be nontoxic, due to the connection with the drinking water treatment process. Rockfos® byproduct should be tested to eliminate the possibility of releasing heavy metals and other hazardous substances to the ground.

3.5. Implications for the practical use of waste materials for P removal

Faced with natural resources limitation, waste materials become an alternative and sustainable substitute in many life and scientific areas. The results of maximum achieved P-adsorption presented in Table 3 and limitation points from Table 6 confirmed increasing interest of the waste materials use in wastewater treatment sector. Unfortunately, a major disadvantage of byproducts reuse is their unstable quality due to variable conditions of the production process.

As regards of performed investigation, calcium rich waste product is exposed to harmful weather conditions during storage and depends on producer prerequisites. DWTRs composition is strictly connected with treated water quality and content of ferric. Nevertheless, the benefits from waste material reuse and possible closing the loop is worth further studies to eliminate unfavourable features. Moreover, waste producers awareness of circular economy may be crucial as constant production process will result with stable byproduct.

Developing legislation for production process and waste condition can oblige to implement best practices to pursue CE, minimize waste and protect the environment (Moktadir et al., 2020a, 2020b). Therefore promising results of performed study should be foundation of legal requirements of production and waste generation process.

4. Conclusions

In summary, this study presents several low-cost or no-cost adsorption materials for the possible application as P-removal substrate. CaO-rich, powder Rockfos® byproduct appears as the most efficient of all investigated materials with maximum theoretical P-sorption capacity calculated from the Langmuir isotherm equal to 256.4 mg/g. Adsorption capacity obtained during the research reached 9.6 mg/g. Assuming the sorption capacity of a commercial product as 100%, using a byproduct we can achieve 105% efficiency with the same material dose. The material with the lowest ability for phosphates binding was DWTRs, where adsorption capacity oscillated near 4.2–4.9 mg/g, which gives about 50% P-removal efficiency following the above assumption. However, considering the results achieved for sorbents with a similar origin and waste character, P-removal ability is sufficient to consider DWTRs as an alternative material to remove phosphorus compounds from wastewater and pursue circular economy assumption.

The Freundlich isotherm fitted an adsorption mechanism for almost all of the tested materials better than others analysed isotherms, with the correlation coefficient and determination coefficient close to 1.0. Furthermore, an assumed pseudo-first and pseudo-second order kinetic

model proved to be well-matched, due to the high values of specific coefficients.

The research performed proved the validity of waste materials and byproducts for phosphates removal according to circular economy requirements. Costs analysis presents a possible opportunity for additional benefits for the waste material/byproduct producers in comparison to the commercial and expensive sorbent. The possibility of P-recovery from these materials after wastewater treatment should be an aim of further studies. The desorption process and toxicity should be carefully investigated, due to potential application as fertiliser components.

Uncited references

(Buhl et al., 1994; Rockfos®, 2020).

CRedit authorship contribution statement

Magda Kasprzyk: Investigation, Data curation, Formal analysis, Writing - original draft, Visualization, Writing - review & editing. **Krzysztof Czerwionka:** Methodology, Investigation, Data curation, Supervision. **Magdalena Gajewska:** Conceptualization, Methodology, Resources, Validation, Supervision.

Declaration of Competing Interest

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

Acknowledgements

This research was co-funded by Norwegian funds, under the Polish-Norwegian Cooperation Research carried out by the National Centre for Research and Development [197025/37/2013].

This paper is a result of financially supported by the project co-financed from European Funds no [POWR.03.05.00-00-Z044/17].

Special thanks to KUFEL Ceramics Company, the sole producer of Rockfos® in Poland, for providing us with research material free of charge.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2020.105335.

Appendix A. Adsorption isotherms

Supplementary data related to this article is attached.

Appendix B. Plot of the linearized form of kinetic models

Supplementary data related to this article is attached.

References

- Acquaye, A., Ibn-Mohammed, T., Genovese, A., Afrifa, G.A., Yamoah, F.A., Oppon, E., 2018. A quantitative model for environmentally sustainable supply chain performance measurement. *Eur. J. Oper. Res.* 269 (1), 188–205. doi:10.1016/j.ejor.2017.10.057.
- Ahmad, T., Ahmad, K., Alam, M., 2016. Sustainable management of water treatment sludge through 3'R' concept. *J. Clean. Prod.* 124, 1–13.
- Al-Ghouti, M.A., Da'ana, D.A., 2020. Guidelines for the use and interpretation of adsorption isotherm models: a review. *J. Hazard. Mater.* 393, 122383. doi:10.1016/j.jhazmat.2020.122383.
- Amann, A., Zoboli, O., Krampe, J., Rechberger, H., Zessner, M., Egle, L., 2018. Environmental impacts of phosphorus recovery from municipal wastewater. *Resour. Conserv. Recycl.* 130, 127–139. doi:10.1016/j.resconrec.2017.11.002.
- Arias, C.A., Brix, H., Johansen, N.H., 2003. Phosphorus removal from municipal wastewater in an experimental two-stage vertical flow constructed wetland system equipped with a calcite filter. *Wat. Sci. Tech.* 48 (5), 51–58.

- Ayawei, N., Ebelegi, A.N., Wankasi, D., 2017. Modelling and interpretation of adsorption isotherms. *J. Chem.* 2017 doi:10.1155/2017/3039817.
- Babatunde, A.O., Zhao, Y.Q., Burke, A.M., Morris, M.A., Hanrahan, J.P., 2009. Characterization of aluminium-based water treatment residual for potential phosphorus removal in engineered wetlands. *Environ. Pollut.* 157, 2830–2836.
- Bai, L., Wang, C., Huang, C., He, L., Pei, Y., 2014. Reuse of drinking water treatment residuals as a substrate in constructed wetlands for sewage tertiary treatment. *Ecol. Eng.* 70, 295–303.
- Berkessa, Y.W., Mereta, S.T., Feyisa, F.F., 2019. Simultaneous removal of nitrate and phosphate from wastewater using solid waste from factory. *Appl. Water Sci.* 9. doi:10.1007/s13201-019-0906-z.
- Brogowski, Z., Renman, G., 2004. Characterization of Opoka as a basis for its use in wastewater treatment. *Polish J. Environ. Stud.* 13 (1), 15–20.
- Buhl, F., Ciba, J., Ciba, L., Górka, P., John, E., Rubel, S., 1994. *Poradnik Chemika analityka*. (eng. Analyst chemist's handbook) Vol. 1.. 1st ed. Wydawnictwo Naukowo-Techniczne, Warsaw 63,66.
- Bus, A., Karczmarczyk, A., 2014. Properties of lime-siliceous rock opoka as reactive material to remove phosphorus from water and wastewater. *Infrastruct. Ecol. Rural Areas* 1 (II), 227–238. doi:10.14597/infraeco.2014.2.1.017.
- Callahan, M.P., Kleinman, P.J.A., Sharpley, A., Stout, W.L., 2002. Assessing the efficacy of alternative phosphorus sorbing soil amendments. *Soil Sci.* 167, 539–547.
- Caniato, F., Caridi, M., Crippa, L., Moretto, A., 2012. Environmental sustainability in fashion supply chains: an exploratory case based research. *Int. J. Prod. Econ.* 135 (2), 659–670. doi:10.1016/j.ijpe.2011.06.001.
- Chiang, Y.W., Ghyselbrecht, K., Santos, R.M., Martens, J.A., Swennen, R., Cappuyns, V., Meesschaert, B., 2012. Adsorption of multi-heavy metals onto water treatment residuals: sorption capacities and applications. *Chem. Eng. J.* 200–202, 405–415.
- Communication from the Commission to the European Parliament, The Council, The European Economic and Social Committee and the Committee of the Regions closing the loop - an EU Action Plan for the Circular Economy, COM/2015/0614 final.
- Copetti, D., Finsterle, K., Marziali, L., Stefani, F., Tartari, G., Douglas, G., Reitzel, K., Spears, B.M., Winfield, L.J., Crosa, G., D'Haese, P., Yesseri, S., Lüring, M., 2015. Eutrophication management in surface waters using lanthanum modified bentonite: a review. *Water Res.* 1–13. doi:10.1016/j.watres.2015.11.056.
- Cornel, P., Schaum, C., 2009. Phosphorus recovery from wastewater: needs, technologies and costs. *Water Sci. Technol.* 59, 1069–1076. doi:10.2166/wst.2009.045.
- Cucarella, V., Renman, G., 2009. Phosphorus sorption capacity of filter materials used for on-site wastewater treatment determined in batch experiments—a comparative study. *J. Environ. Qual.* 38 (2), 381. doi:10.2134/jeq2008.0192.
- Cucarella, V., Zaleski, T., Mazurek, R., Renman, G., 2007. Fertilizer potential of calcium-rich substrates used for phosphorus removal from wastewater. *Polish J. Environ. Stud.* 16 (6), 817–822.
- Dayton, E.A., Basta, N.T., 2005. Use of drinking water treatment residuals as a potential best management practice to reduce phosphorus risk index scores. *J. Environ. Qual.* 34, 2112–2117.
- Del Bubba, M., Arias, C.A., Brix, H., 2003. Phosphorus adsorption maximum of sands for use as media in subsurface flow constructed reed beds as measured by the Langmuir isotherm. *Water Res.* 37 (14), 3390–3400. doi:10.1016/S0043-1354(03)00231-8.
- Douglas, G.B., Adeney, J.A., Robb, M., 1999. A novel technique for reducing bioavailable phosphorus in water and sediments. *Int. Assoc. Water Qual. Conf. Diffuse Pollut.* 517–523.
- Douglas, G.B., Lüring, M., Spears, B.M., 2016. Assessment of changes in potential nutrient limitation in an impounded river after application of lanthanum-modified bentonite. *Water Res.* 1–8. doi:10.1016/j.watres.2016.02.005.
- Drizo, A., Forget, C., Chapuis, R.P., Comeau, Y., 2006. Phosphorus removal by electric arc furnace steel slag and serpentine. *Water Res.* 40, 1547–1554. doi:10.1016/j.watres.2006.02.001.
- Drizo, A., Frost, C.A., Grace, J., Smith, K.A., 1999. Physico-chemical screening of phosphate-removing substrates for use in constructed wetland systems. *Water Res.* 33 (17), 3595–3602. doi:10.1016/S0043-1354(99)00082-2.
- Ebadi, A., Soltan, J.S., 2009. What is the correct form of BET isotherm for modeling liquid phase adsorption?, 65–73. 10.1007/s10450-009-9151-3.
- Egle, L., Rechberger, H., Krampe, J., Zessner, M., 2016. Phosphorus recovery from municipal wastewater: an integrated comparative technological, environmental and economic assessment of P recovery technologies. *Sci. Total Environ.* 571, 522–542. doi:10.1016/j.scitotenv.2016.07.019.
- Egle, L., Rechberger, H., Zessner, M., 2015. Overview and description of technologies for recovering phosphorus from municipal wastewater. *Resour. Conserv. Recycl.* 105, 325–346. doi:10.1016/j.resconrec.2015.09.016.
- Febrianto, J., Kosasih, A.N., Sunarso, J., Ju, Y.H., Indraswati, N., Ismadji, S., 2009. Equilibrium and kinetic studies in adsorption of heavy metals using biosorbent: a summary of recent studies. *J. Hazard. Mater.* 162, 616–645. doi:10.1016/j.jhazmat.2008.06.042.
- Gobin, O.C., 2006. Thesis. SBA-16 Materials Synthesis, Diffusion and Sorption Properties. Laval University, Ste-Foy, Quebec, Canada.
- Groves, S., 2010. Eco-toxicity Assessment of Phoslock. PWS Report Number: TR 022/09. Phoslock Water Solutions Limited.
- Guerra-Rodríguez, S., Oulego, P., Rodríguez, E., Singh, D.N., Rodríguez-Chueca, J., 2020. Towards the implementation of circular economy in the wastewater sector: challenges and opportunities. *Water (Switzerland)* 12, 1–57. doi:10.3390/w12051431.
- Haghseresh, F., 2006. A revolution in phosphorus removal. PWS, Report Number PS-06, (October), 22. Retrieved from http://www.klamathbasinacrisis.org/algae/B1_PhoslockDescriptionFHOct2006.pdf.
- Haghseresh, F., Wang, S., Do, D.D., 2009. A novel lanthanum-modified bentonite, Phoslock®, for phosphate removal from wastewaters. *Appl. Clay Sci.* 46, 369–375.

- Han, R., Zhang, J., Zou, W., Shi, J., Liu, H., 2005. Equilibrium biosorption isotherm for lead ion on chaff. *J. Hazard. Mater.* 125, 266–271. doi:10.1016/j.jhazmat.2005.05.031.
- Ho, Y.S., Chiang, C.C., 2001. Sorption studies of acid dye by mixed sorbents. *Adsorption* 7 (2), 139–147. doi:10.1023/A:1011652224816.
- Ho, Y.S., McKay, G., 1998. Kinetic models for the sorption of dye from aqueous solution by wood. *Process Saf. Environ. Protect.* 76 (2), 183–191. doi:10.1205/095758298529326.
- Ho, Y.S., McKay, G., 1999. Pseudo-second order model for sorption processes. *Process Biochem* 34, 451–465.
- Hossain, N., Bhuiyan, M.A., Pramanik, B.K., Nizamuddin, S., Griffin, G., 2020. Waste materials for wastewater treatment and waste adsorbents for biofuel and cement supplement applications: a critical review. *J. Clean. Prod.* 255. doi:10.1016/j.jclepro.2020.120261.
- Hu, Y.S., Zhao, Y.Q., Sorohan, B., 2011. Removal of glyphosate from aqueous environment by adsorption using water industrial residual. *Desalination* 271, 150–156.
- Huang, W., Yu, X., Tang, J., Zhu, Y., Zhang, Y., Li, D., 2015. Enhanced adsorption of phosphate by flower-like mesoporous silica spheres loaded with lanthanum. *Microporous Mesoporous Mater.* 217, 225–232. doi:10.1016/j.micromeso.2015.06.031.
- Hukari, S., Hermann, L., Nattorp, A., 2016. From wastewater to fertilisers - technical overview and critical review of European legislation governing phosphorus recycling. *Sci. Total Environ.* 542, 1127–1135. doi:10.1016/j.scitotenv.2015.09.064.
- Ippolito, J.A., Barbarick, K.A., Elliott, H.A., 2011. Drinking water treatment residuals: a review of recent uses. *J. Environ. Qual.* 40, 1–12.
- Izidor, J.C., Kim, M.C., Belleli, V.F., Pane, M.C., Botelho Junior, A.B., Espinosa, D.C.R., Tenório, J.A.S., 2019. Synthesis of zeolite A using the waste of iron mine tailings dam and its application for industrial effluent treatment. *J. Sustain. Min.* 18, 277–286. doi:10.1016/j.jsm.2019.11.001.
- Johansson Westholm, L., 2006. Substrates for phosphorus removal e potential benefits for on-site wastewater treatment? *Water Res.* 40 (1), 23–36. doi:10.1016/j.watres.2005.11.006.
- Kadlec, R.H., Wallace, S., 2009. *Treatment Wetlands*. 2nd ed. CRC Press Taylor & Francis Group, Boca Raton, London, New York.
- Kasprzyk, M., Gajewska, M., 2019. Phosphorus removal by application of natural and semi-natural materials for possible recovery according to assumptions of circular economy and closed circuit of P. *Sci. Total Environ.* 650, 249–256. doi:10.1016/j.scitotenv.2018.09.034.
- Kasprzyk, M., Obarska-Pempkowiak, H., Masi, F., Gajewska, M., 2018. Possibilities of Phoslock® application to remove phosphorus compounds from wastewater treated in hybrid wetlands. *Ecol. Eng.* 122, 84–90. doi:10.1016/j.ecoleng.2018.07.020.
- Leader, J.W., Reddy, K.R., Wilkie, A.C., 2005. Optimization of low-cost phosphorus removal from wastewater using co-treatments with constructed wetlands. *Water Sci. Technol.* 51 (9), 283–290. doi:10.2166/wst.2005.0338.
- Leader, W.J., Dunne, E.J., Reddy, K.R., 2008. Phosphorus sorbing materials: sorption dynamics and physicochemical characteristics. *J. Environ. Qual.* 37 (1), 174–181. doi:10.2134/jeq2007.0148.
- Leyva-Díaz, J.C., Monteoliva-García, A., Martín-Pascual, J., Munio, M.M., García-Mesa, J.J., Poyatos, J.M., 2020. Moving bed biofilm reactor as an alternative wastewater treatment process for nutrient removal and recovery in the circular economy model. *Bioresour. Technol.* 299, 122631. doi:10.1016/j.biortech.2019.122631.
- Li, X., Cui, J., Pei, Y., 2018. Granulation of drinking water treatment residuals as applicable media for phosphorus removal. *J. Environ. Manage.* 213, 36–46. doi:10.1016/j.jenvman.2018.02.056.
- Li, X., Yu, D., Su, L., Pei, Y., 2019. Facile method to granulate drinking water treatment residues as a potential media for phosphate removal. *Colloids Surfaces A* press doi:10.1016/j.colsurfa.2019.124198.
- Lin, L., Xu, X., Papelis, C., Cath, T.Y., Xu, P., 2014. Sorption of metals and metalloids from reverse osmosis concentrate on drinking water treatment solids. *Sep. Purif. Technol.* 134, 37–45.
- Liu, X., Zhang, L., 2015. Removal of phosphate anions using the modified chitosan beads: adsorption kinetic, isotherm and mechanism studies. *Powder Technol* 277, 112–119. doi:10.1016/j.powtec.2015.02.055.
- Moktadir, M.A., Ahmadi, H.B., Sultana, R., Zohra, F.T., Liou, J.J.H., Rezaei, J., 2020a. Circular economy practices in the leather industry: a practical step towards sustainable development. *J. Clean. Prod.* 251. doi:10.1016/j.jclepro.2019.119737.
- Moktadir, M.A., Kumar, A., Ali, S.M., Paul, S.K., Sultana, R., Rezaei, J., 2020b. Critical success factors for a circular economy: implications for business strategy and the environment. *Bus. Strateg. Environ.* 0, 1–25. doi:10.1002/bse.2600.
- Nagar, R., Sarkar, D., Makris, K.C., Datta, R., 2010. Effect of solution chemistry on arsenic sorption by Fe- and Al-based drinking-water treatment residuals. *Chemosphere* 78, 1289–1305.
- Nastawny, M., Jucherski, A., Walczowski, A., Józwiakowski, K., Pytka, A., Gizińska-Górna, M., Marzec, M., Gajewska, M., Marczuk, A., Zarajczyk, J., 2015. Preliminary evaluation of selected mineral adsorbents used to remove phosphorus from domestic wastewater. *Przem. Chem.* 10 (94), 1000–1004 Polish doi:10.15199/62.2015.10.XX.
- Penn, C.J., Bryant, R.B., 2006. Application of phosphorus sorbing materials to streamside cattle loafing areas. *J. Soil Water Conserv.* 61, 303–309.
- Perez, I.D., Botelho Junior, A.B., Aliprandini, P., Espinosa, D.C.R., 2020. Copper recovery from nickel laterite with high-iron content: a continuous process from mining waste. *Can. J. Chem. Eng.* 98, 957–968. doi:10.1002/cjce.23667.
- Prasanna Kumar, Y., King, P., Prasad, V.S.R.K., 2006. Equilibrium and kinetic studies for the biosorption system of copper(II) ion from aqueous solution using *Tectona grandis* L.f. leaves powder. *J. Hazard. Mater.* 137, 1211–1217. doi:10.1016/j.jhazmat.2006.04.006.
- Pratt, C., Shilton, A., 2010. Active slag filters—simple and sustainable phosphorus removal from wastewater using steel industry byproduct. *Water Sci. Technol.* 62 (8), 1713–1718. doi:10.2166/wst.2010.389.
- Rockfos® Product Data Sheet. 2020 Retrieved from <http://www.ceramika-kufel.pl/rockfos/>.
- Ross, G., Haghseresh, F., Cloete, T.E., 2008. The effect of pH and anoxia on the performance of Phoslock, a phosphorus binding clay. *Harmful Algae* 7 (4), 545–550. doi:10.1016/j.hal.2007.12.007.
- Sfez, S., De Meester, S., Vlaeminck, S.E., Dewulf, J., 2019. Improving the resource footprint evaluation of products recovered from wastewater: a discussion on appropriate allocation in the context of circular economy. *Resour. Conserv. Recycl.* 148, 132–144. doi:10.1016/j.resconrec.2019.03.029.
- Singh, N.B., Nagpal, G., Agrawal, S., Rachna, 2018. Water purification by using Adsorbents: a Review. *Environ. Technol. Innov.* 11, 187–240. doi:10.1016/j.eti.2018.05.006.
- Smith, V.H., Tilman, G.D., Nekola, J.C., 1999. Eutrophication: impacts of excess nutrient inputs on freshwater, marine, and terrestrial ecosystems. *Environ. Pollution* 100, 179–196. doi:10.1016/S0269-7491(99)00091-3.
- Song, X., Pan, Y., Wu, Q., Cheng, Z., Ma, W., 2011. Phosphate removal from aqueous solutions by adsorption using ferric sludge. *Desalination* 280 (1–3), 384–390. doi:10.1016/j.desal.2011.07.028.
- Tan, Y., Haver, J.H.O. 2004. Use of the BET adsorption isotherm equation to examine styrene adsorbilization by nonionic surfactants at the water – silica interface, 279, 289–295. doi:10.1016/j.jcis.2004.06.101.
- van Oosterhout, F., Goitom, E., Roessink, I., Lüring, M., 2014. Lanthanum from a modified clay used in eutrophication control is bioavailable to the marbled crayfish (*Procambarus fallax f. virginalis*). *PLoS ONE* 9 (7), e102410. doi:10.1371/journal.pone.0102410.
- van Oosterhout, F., Lüring, M., 2013. The effect of phosphorus binding clay (Phoslock®) in mitigating cyanobacterial nuisance: a laboratory study on the effects on water quality variables and plankton. *Hydrobiologia* 710, 265–277. doi:10.1007/s10750-012-1206-x.
- Vohla, C., Köiv, M., Bavor, H.J., Chazarenc, F., Mander, Ü, 2011. Filter materials for phosphorus removal from wastewater in treatment wetlands — a review. *Ecol. Eng.* 37, 70–89. doi:10.1016/j.ecoleng.2009.08.003.
- Vymazal, J., 2011. Constructed wetlands for wastewater treatment: five decades of experience. *Environ. Sci. Technol.* 45, 61–69.
- Wang, C., Guo, W., Tian, B., Pei, Y., Zhang, K., 2011. Characteristics and kinetics of phosphate adsorption on dewatered ferric-alum residuals. *J. Environ. Sci. Health A* 46, 1632–1639.
- Wang, C., Liu, J., Pei, Y., 2013. Effect of hydrogen sulfide on phosphorus lability in lake sediments amended with drinking water treatment residuals. *Chemosphere* 91, 1344–1348.
- Wang, C., Wu, Y., Bai, L., Zhao, Y., Yan, Z., Jiang, H., Liu, X., 2018. Recycling of drinking water treatment residue as an additional medium in columns for effective P removal from eutrophic surface water, 217, 363–372. doi:10.1016/j.jenvman.2018.03.128.
- Wang, Z., Shen, D., Shen, F., Li, T., 2016. Phosphate adsorption on lanthanum loaded biochar. *Chemosphere* 150, 1–7. doi:10.1016/j.chemosphere.2016.02.004.
- Xu, Z., Cai, J., Pan, B., 2013. Mathematically modeling fixed-bed adsorption in aqueous systems *, 14(3), 155–176. doi:10.1631/jzus.A1300029.
- Ye, Y., Ngo, H.H., Guo, W., Liu, Y., Li, J., Liu, Y., Jia, H., 2017. Insight into chemical phosphate recovery from municipal wastewater. *Sci. Total Environ.* 576, 159–171. doi:10.1016/j.scitotenv.2016.10.078.
- Zamparas, M., Gianni, A., Stathi, P., Deligiannakis, Y., Zacharias, I., 2012. Removal of phosphate from natural waters using innovative modified bentonites. *Appl. Clay Sci.* 62–63, 101–106.
- Zamparas, M., Kyriakopoulos, G.L., Drosos, M., Kapsalis, V.C., Kalavrouziotis, I.K., 2020. Novel composite materials for lake restoration: a new approach impacting on ecology and circular economy. *Sustainability* 12, 1–17 20203397.
- Zhou, Y.F., Haynes, R.J., 2011. Removal of Pb(II), Cr(III) and Cr(VI) from aqueous solutions using alum-derived water treatment sludge. *Water Air Soil Poll.* 215, 631–643.
- Zhu, T., Maehlum, T., Jenssen, P.D., Krogstad, T., 2003. Phosphorus sorption characteristics of a light-weight aggregate. *Wat. Sci. Tech.* 48 (5), 93–100.