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Evaluation of the sustainability of technologies to recover phosphorus from sewage sludge ash based on embodied energy and CO2 footprint

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Abstract

This paper reports a novel and simplified approach for sustainability evaluation of new technologies, based on the use of two parameters (i.e. embodied energy and CO2 footprint) that account for the energy and emissions involved in the formation of a material, suitable to perform pre-screening analysis, preliminary to life cycle assessment. This new approach is presented and used to compare the available technologies for phosphorous recovery from sewage sludge ash. A dimensionless index, defined as SUBRAW index, is used to compare the results about the environmental impact of each selected substituting material/process. This method is applied to full, pilot, and laboratory scale processes available in literature in the context of phosphorus extraction technologies. The results show that wet chemical leaching approaches appear to be the most sustainable methods.

1. Introduction

Phosphorus (P) is a strategic resource, being an irreplaceable macronutrient for crops, cells, humans and animals. In particular, by 2050 the P production is expected to increase by 70% overall, in order to satisfy the rapidly growing P request (Cieslik and Konieczka, 2017). On the other hand, there is a very high degree of uncertainty as to the correspondence of the estimates of remaining Pore deposits in years with actual ultimately recoverable phosphate resources: 50e100 years (Cordell and White, 2011), 400e500 years (Van Kauwenbergh, 2010), and more than 1000 years (Mew, 2016; Reijnders, 2014; Scholz and Wellmer, 2013; Van Vuuren et al., 2010). In face of the actual human population growth phosphate rock is a mandatory material in the production of fertilisers such as single superphosphate (SSP), diammonium phosphate (DAP), dicalcium phosphate (DCP) and magnesium ammonium phosphate (MAP). This process is preceded by phosphate rocks exploitation and beneficiation to concentrate P-ore, which is then acid-leached to extract P and produce the fertilizers. Then, the need of P-rich fertilisers prompts a significant concern on the proper management of this resource (Li et al., 2019).

The European Union has a scarce supply of phosphate rocks deposits (Fig. 1), implying the inevitable import of P from other countries as China, Morocco, Russia, Algeria and the USA (Ptacek, 2016; Walan et al., 2014): the supply risk could lead to geopolitical problems for companies and governments in terms of phosphate rocks mining costs, physical adequacy of phosphate rocks resources and economic access (Mew, 2016). Thus, phosphate rocks were added to the list of the 20 Critical Raw Materials (European Commission, 2014) and policies to assess P recovering processes were promoted (Egle et al., 2016). In this frame, it is urgent to

encourage the development of environmentally sustainable recovery technologies (Szogi and Vanotti, 2009). In particular, organic waste streams contain a considerable amount of P in various organic and inorganic compounds.

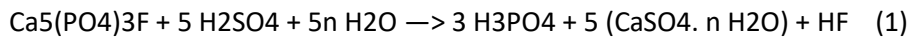
Among the waste streams, sewage sludge is one of the most promising sources of P especially in terms of percentage of produced waste (Udaeta et al., 2018). In addition, excess of P could be a problem in the wastewater flow because it could lead to algal blooms and eutrophication of ponds and lakes (Vanotti et al., 2003). Therefore, the European Water Framework Directive in 2000/60/EC (European Commission, 2000) claimed that the maximum allowable concentration of P in wastewater treatment plant effluents must be lower than 0.15 mg P/dm³.

Among the available organic waste management technologies, incineration is able to reduce sewage sludge volume up to 80% and to eliminate pathogens as well as toxic organic substances (Donatello and Cheeseman, 2013). Moreover, the incineration could potentially increase up to about five times (depending on the combustion system) the P percentage in the residue, which makes this material comparable with low grade phosphate rocks with a P concentration around 8% (Cieslik and Konieczka, 2017; Franz, 2008). The recovery of P from sewage sludge ash has the additional advantage to eventually decrease the consumption of natural resources and the environmental and economic impact related with phosphate rocks mining (Pasquali et al., 2018) in case it would be integrated with already established P extraction processes from the primary deposits (Langeveld, 2018). Several strategies were proposed for the P recovering (Tan and Lagerkvist, 2011) and several Life Cycle Assessment (LCA) studies addressed to different treatments of wastes containing P (as incineration, land spreading, composting, wet oxidation, and landfill) have been performed (Lederer and Rechberger, 2010; Lombardi et al., 2017; Tarpani et al., 2020). However, a global and straightforward study is needed to compare the technologies available in literature and evaluate their economic and environmental sustainability. In this paper, we proposed a new approach to directly evaluate the sustainability of the technologies to recovery P derived from secondary waste streams (i.e. sewage sludge ash (SSA)). In this study full, pilot, and laboratory-scale technologies were considered. The novel approach is based on a simplified index, defined as SUB-RAW index, that has been applied to quantify the sustainability of the recovering technologies in comparison to mining. The SUB-RAW index is based on two parameters: "CO₂ footprint" (CF) and the "embodied energy" (EE). CF gives an inherent value of gas emissions expressed as CO₂ equivalent, which is an indicator of the environmental impact, while EE is related to energy consumption. SUB-RAW index can be used to quantify the advantage of P-recovery technologies (Bontempi, 2017a, 2017b; Pasquali et al., 2018). The proposed approach does not require the knowledge of all the stages of a material life cycle. As a consequence, it allows to obtain a parameter, that is more rapid and easy-to-calculate in comparison to indicators that could be obtained by using LCA. In addition, it also allows to evaluate processes developed only at laboratory-scale. This makes the new proposed procedure useful for the screening of novel technologies and to support the evaluation of early-stage technologies sustainability.

2. Methods

2.1. The phosphorous extraction

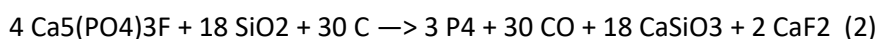
Among the phosphate rocks (carbonate apatite, fluorapatite, hydroxyapatite and sulphur-apatite) the fluorapatite (Ca₅(PO₄)₃F) from sedimentary rock deposits has been so far of major commercial importance since it has low reactivity because of the high crystallinity (Samreen and Kausar, 2019). Therefore, the fluorapatite was chosen in this work as the reference for the evaluation of P extraction cost. Most commonly finely grounded phosphate rock is leached with sulphuric acid, but other strong acids such as hydrochloric acid and nitric acid are also used. The simplified reaction involving sulphuric acid as leaching agent proceeds according to the following equation:



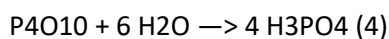
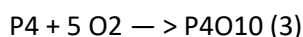
The purity of the products (phosphoric acid (H₃PO₄; PA) and phosphor-gypsum) rely on the quality of phosphate rock (Mochiyama, 2018). Because the purity of the products depends on the phosphate rock quality, it will not be considered in our analysis. In comparison to phosphate rocks, phosphate from SSA is present in mixed oxides with Ca, Mg, Fe and Al, and as calcium phosphate whitlockite (Ca₉(PO₄)₆PO₃OH), whereas the former P is mainly hosted by apatite, which contains chlorides, fluorides, carbonates and hydroxides that may affect the efficiency of acidulation process (Donatello and Cheeseman, 2013; Langeveld, 2018). However, the Ca, Mg, Fe, Al concentrations depend on the sewage sludge type, the combustion system, and the incineration conditions (Donatello and Cheeseman, 2013). Moreover, SSA is powdered and the milling is not necessary, while phosphate rock the sludge must be filtered to separate calcium sulphate crystals (phosphor-gypsum) and PA.

Another process leading to (pure) PA is based on the dry thermo-reductive process. In this case, the product is obtained by the oxidation of white phosphorus (P₄), generated by the carbothermal reduction of phosphate rock through the Wohler process, in which apatite reacts with coke (reducing agent) and silica (as flint pebbles) in low shaft electric furnace at 1200-1500 °C. The resulting product (P₄) is very pure, but the environmental impacts of the global process in terms of energy consumption and gaseous emissions is high and needs access to locally sourced and low cost Acronyms P phosphorus SSA sewage sludge ash PA phosphoric acid P₄ white phosphorus CF CO₂ footprint EE embodied energy Fig. 1. Overview of the main phosphate rocks mining areas (data from (Granta Design, 2019)). The table on the right of the figure shows the % P₂O₅ region by region (data from (Ptacek, 2016)). A. Fahimi, S. Federici, L.E. Depero et al. Journal of Cleaner Production 289 (2021) 125762 2 electricity, coal and phosphate rock (de Boer et al., 2018; Diskowski and Hofmann, 2000; Mochiyama, 2018).

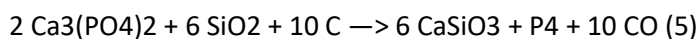
The reaction, in the case of phosphate rocks, is the following (de Boer et al., 2018):



After, the oxidation of P₄ followed by hydrolysis is necessary:



For SSA, a thermal process based on inductive heating up to 1600 °C under reducing environment has been recently proposed (Langeveld, 2018):



However, the amount of SiO₂ in SSA is lower than in phosphate rocks (Ptacek, 2016; Udaeta et al., 2018), and the temperature required for the reaction is higher (Zhang et al., 2018). Therefore, different energy consumptions and emissions are expected for the processes of obtaining P from the natural rocks and SSA.

2.2. ESCAPE approach

ESCAPE (Evaluation of Sustainability of material substitution using CARbon footPrint by a simplified approach) is a novel method introduced by Bontempi (Bontempi, 2017b, 2017c, Ducoli et al., submitted). It

is based on the SUB-RAW index, a simple, direct, and versatile index, which allows the environmental impact evaluation of waste or by-product materials if used in substitution of natural resources. In this work, ESCAPE is applied for the environmental assessment of P-recovery technologies involving SSA as potential starting secondary raw material.

SUB-RAW index considers two parameters strongly correlated (see Fig. 2) (Bontempi, 2017a,d): 1) the EE that includes the energies directly and indirectly needed for the production of 1 kg of the material; 2) the CF that represents the equivalent mass of greenhouse gases released into the atmosphere when 1 kg of the material is produced (equivalent kg of CO₂). The parameters are calculated in MJ and Kg CO₂ respectively. They can be normalized to a reference system defined arbitrarily, and in this study the EE and CF are normalized to 1 kg of P produced (kg(P)). The EE and CF values of the reagents used in the P-extraction technologies were reported in several databases and in this work CES Selector 2019 (Granta Design, 2019) and openLCA (GreenDelta, 2006) with their databases (commercial Ecoinvent v. 2.2 and free Ecoinvent v. 3.3 respectively) were used. The “Eco Audit Tool” of the CES Selector 2019 combines user-defined input with EE and CF values, processing energy and transport type to create the energy breakdown (Ashby et al., 2009). This tool was used for the calculation of EE and CF associated to the total operating power consumption (expressed in Watt, W) for moving mechanical components (mixers, crushers, mills, vacuum filters etc.) or heating (for thermal annealing). The EE and CF parameters were evaluated for each step of the considered technology, and the SUB-RAW index was calculated by the formula (Bontempi, 2017b):

$$\text{SUB-RAW index} = [\log(\text{EERAW}/(\text{MJ}/\text{kg})) - \log(\text{EESUB}/(\text{MJ}/\text{kg})) + \log(\text{CFRAW}) - \log(\text{CFSUB})]/2 \quad (6)$$

EERAW and CFRAW are the EE (MJ/kg(P)) and CF (kg CO₂/kg(P)) of the reference process, i.e. the P-extraction from the phosphate rocks, while EESUB and CFSUB are the EE (MJ/kg(P)) and CF (kg CO₂/kg(P)) of the P-extraction process from the SSA. The logarithm in the formula allows having a direct and simple comparison taking into account an average of the environmental emission impact and energy consumed. Therefore, the SUB-RAW index is a dimensionless number (both EERAW and EESUB were divided by MJ/kg to make SUB-RAW index dimensionless) indicating the level of sustainability of a recovery process. If negative, the recovery process is less sustainable in comparison to P-extraction from the phosphate rocks.

The calculations are carried out for the published full/pilot/ laboratory-scale technologies listed in Table 1. The necessary data for the calculation were input mass flow of reagents, input mass flow of raw material/waste material, the setting of electrical components and their relative operating power consumption. A categorisation of the technologies (Table 1), based on the availability of all the data source, has been done to define a repeatable pattern for the process flowsheet. The processes reporting all the necessary information were marked with * in Table 1. When all the necessary input data for the ESCAPE evaluation were not available, plausible hypothesis were made by the authors to take into account all the steps of the process. On the basis of the amount of available data, when few or several assumptions were necessary to perform the SUB-RAW index calculation, the technologies were marked with **, and with*** respectively. A file describing all the evaluated steps and the eventual introduced hypothesis is available as supporting information (S1). Usually, resource use and chemical demand for the P-recovery processes can be transferred from laboratory/pilot scale to full scale using useful relations while electricity demand and heat management have to be estimated. Therefore, the assumptions done in the calculations were the following:

- The process was considered a steady state unidirectional flowsheet. Thus, the model does not provide any temporal (constant mass flow of reagents) or spatial (no distinction of close or open environment) resolution of emissions. Besides, input data only regard reagents, raw material/waste material, and electricity directly applied to the mainline of the process (e.g. downstream contribution of output materials are not considered).
- Every leaching/neutralisation/precipitation step being part of a wet chemical leaching process was carried out in standard conditions (T = 25 °C, P = 1 bar).

- EE and CF to produce SSA were assumed to be equal to 0 MJ/ kg(ash) and 0 kgCO₂/kg(ash) since the material is a residue. EE and CF for phosphate rocks were taken from the database (Rodríguez, 2016) used through OpenLCA software (GreenDelta, 2006) and are equal to 7.77 MJ/kg(P) and 0.62 kg CO₂/kg(P) respectively.
- Circulating processed water was considered, and its EE and CF are assumed to be 0.01 MJ/kg(H₂O) and 0 kg CO₂/kg(H₂O) (Mo et al., 2011). Using commercially available immersion pumps, which is based on the water volume flow in the process, the EE and CF calculated using CES Selector 2019 confirmed the values.
- SSA and phosphate rocks composition and other boundary conditions can vary on a broader range. For example, Franz (2008) reported that total P concentration (%) in SSA varies between 4% and 9% (that means in terms of P₂O₅: 9.2% and 20.6%), whereas in phosphate rocks it varies between 7.9% and 17.5% (in terms of P₂O₅: 18.1% and 40.1%) (Hore-Lacy, 2016), which can impact on the efficiency of the considered process. Thus, considering only the common range about P concentration, its mean value was assumed to be equal to 8.45% (total amount of P) for both SSA and phosphate rocks in order to make them comparable (Cieślak and Konieczka, 2017; de Boer et al., 2018; Ottosen et al., 2013; Weigand et al., 2013).
- The flowsheet of wet process ended up with a chemical solution where P is under various soluble compounds (e.g. PA at different grades, calcium phosphate, aluminium phosphate, etc.). In the dry process P₄ is obtained, no further oxidation step was considered aimed to obtain high purity PA.

2.3. Model validation

In order to have a basis for a suitable comparison among different processes, the proposed approach was validated. The wet chemical leaching process using sulphuric acid for the P-extraction from phosphate rocks, labelled as “Rhone-Poulenc ^ ” process (Hutchins, 1993), was chosen as the reference technology (Cheremisinoff, 1995; Gilmour, 2013; Hocking, 1985), because over 80% of P is led to obtaining fertilisers (Schnee and Opitz, 2018) and dry processes mostly provide high-grade PA used for food and industrial applications (de Boer et al., 2018).

Fig. 3 shows the EE and CF parameters of the reference process and the reference values provided by the CES Selector 2019 (Granta Design, 2019). In particular, the reference process (marked with “1” in Fig. 3) evaluated by ESCAPE has as major contribution the use of sulphuric acid, i.e. over 80% of emissions, with 0.84 kg/kg(ore) (emission values of sulphuric acid in OpenLCA are EE = 7.355 MJ/ kg(sulphuric acid); CF = 0.21 kg CO₂/kg (sulphuric acid)). This contribution was added to the emissions of the phosphate rocks as starting mined and grounded material (EE = 7.77 MJ/kg(P) and CF = 0.62 kg CO₂/kg(P)) and the working electrical facilities (e.g. tank mixers, belt conveyors, vacuum dry filter, pan filter, single effect vaporiser, etc). This contribution depends on the size of the working area, mostly regulated by the input mass flow of raw material entering the process. In this study, the capacity is referred to an instantaneous operating power consumption equal to 250 kW, including a generated steam for the vaporisation of PA (concentrated up to 70%) of 0.285 kg/kg(ore). The calculated values of EE and CF are 0.85 MJ/kg(ore) and 0.05 kg CO₂/kg(ore), respectively. Finally, it was added the oil drying contribution of 0.17 kWh/ kg(ore), taken from (Egle, 2016) in which it was actually applied to a flowsheet of another similar process (Weigand et al., 2013), corresponding to EE = 1.33 MJ/kg(ore) and CF = 0.08 kg CO₂/ kg(ore).

Since phosphate rocks containing 19.36% of P₂O₅ (8.45% of P) were considered in this study, the obtained emissions parameters equal to EE = 102.32 MJ/kg(P) and CF = 4 kg CO₂/kg(P) (marked with “1” in Fig. 3). The CES Selector provided values of the extraction of pure P₄, and the arrow marked domain in Fig. 3 represents different dry processes applied to different phosphate rocks. CES Selector domain (EE = 218e240 MJ/kg(P); CF = 9.52e10.5 kg CO₂/kg(P)) represents the emissions values for all the dry processes, taken from the Ecoinvent v. 2.2 database (Frischknecht et al., 2007). The difference between these values and that calculated

by ESCAPE was due to the high temperature (1200e1500 °C) of the dry process needed to obtain P4. To compare the results, a further step in this study model was considered, after assuming to obtain PA, to thermally reduce the PA. Besides, taking into consideration the purification step. Indeed, phosphate rocks may contain impurities such as As, Cd, U and Cd removal alone from PA leads to increase the EE of the process by 32 MJ/kg(P) (Pasquali et al., 2018). Thus, this contribution was added even if not all the phosphate rocks require this treatment (marked with “2” in Fig. 3). The thermal reduction to obtain P4 uses a 50 MW electrical power consumption furnace (the size depends on the materials input flow) (Diskowski and Hofmann, 2000) in which PA undergoes to the electric to mechanical power consumption of 5.85 kWh/kg(P) and electric to thermal power consumption of 5.2 kWh/kg(P). The first value accounted only for the energy consumption of the chemical reactions and corresponds to EE = 51.6 MJ/kg(P) and CF = 3.1 kg CO₂/ kg(P), meanwhile the second one accounted for heat content of slag and furnace gases and corresponds to EE = 40.81 MJ/kg(P) and CF = 2.45 kg CO₂/kg(P). The calculation did not consider the contribution to EE and CF of the reducing reagents, C and SiO₂. Adding these contributions (marked with “3” in Fig. 3), the value of the reference process (EE = 226.76 MJ/kg(P) and CF = 12.75 kg CO₂/ kg(P)) was similar to those found by CES Selector 2019. These results demonstrate the reliability of ESCAPE approach in evaluating the EE and CF of P-extraction technologies. However, it should be remarked that the results were subject to a justified uncertainty explained by the multiple assumptions, along with all the procedures to calculate the emissions of reagents collected inside the Ecoinvent database.

3. Results

ESCAPE was applied to evaluate the EE and CF of the technologies reported in Table 1, the results are reported in Fig. 4, and all the steps and corresponding evaluated values are shown in Fig. 4, which is reported in supporting information (S1)

To show the process steps, as a single example, the application of ESCAPE to the Lotus Project technology (Nakagawa and Ohta, 2018) was reported in detail. This technology is one of those processes which data were fully provided (marked with * in Table 1), meaning that no assumptions were necessary to provide the sustainability evaluation. This case was particularly described here since it is a wet chemical leaching process using alkaline chemicals as leaching agent, whereas the majority of the wet processes are based on acid-leaching.

The Lotus Project is a full-scale process for recovering P from SSA, which is under operation in Gifu City (Japan; Fig. 5). The plant currently generates 200 tons/year of P recovered as hydroxyapatite. The P-extraction process is based on wet alkaline-leaching technology by adding sodium hydroxide to SSA (NaOH(4%), 0.121 kg/ kg(ash)) at 50e70 °C for 1.5 h (the thermal energy is provided by the residual heat of the incinerator, which was neglected in the present work). The process description provides the information to evaluate the global amount of corresponding EE and CF. Moreover, based on the assumption of a steady-state mass flow, calculations assumed the process in a continuous flow. In the second step, the leachate is obtained using a membrane, and then the solid residue is washed with sulphuric acid (H₂SO₄(98%), 0.04 kg/kg(ash)) for 5e30 min at 50e70 °C. The calculation of the by-product management and the heavy metal treatment (in the waste) were (solely in this validation case) considered part of the flowsheet (De-P-ash). Since the emissions data from Ecoinvent database were provided for reagents at their purest state, the contribution of water in the diluted reagent solutions were considered separately. The leachate, containing AlPO₄, precipitates as hydroxyapatite by adding hydrated lime (Ca(OH)₂, 0.431 kg/kg(ash)) at 20e50 °C for 9 h.

In the final step, the precipitated hydroxyapatite is washed with water, filtered, dried and pelletized, meanwhile the residual liquid from the precipitation tank is returned to the head of leaching reactor. This step was not considered in the calculations since we considered a unidirectional process (see Section 2). The

water for washing the hydroxyapatite outside the neutralisation tank and diluting concentrated reagents was quantified to 3 kg/kg(ash) as reported in (Egle, 2016). This gives an overall value for the EE equal to 0.03 MJ/kg(ash) for water contribution, which was not relevant for the overall process. The electricity consumption necessary to move mixers in leaching, neutralisation and precipitation tank, membrane-like solid-liquid separator, drying furnace, pelletizer and the resulting sum of instantaneous operating power consumption resulted equal to 36 kW. This value was input in Eco Audit Tool, obtaining a yearly value for each emissions parameter. Then, considering the yearly SSA mass flow, EE and CF were equal to 0.8 MJ/kg(ash) and 0.07 kg CO₂/kg(ash), respectively.

The contribution of the reagents, namely sodium hydroxide (0.12 MJ/kg(ash), 0.001 kg CO₂/kg(ash)), sulphuric acid (0.29 MJ/kg(ash), 0.01 kg CO₂/kg(ash)), hydrated lime (1.65 MJ/kg(ash), 0.39 kg CO₂/kg(ash)), and water (0.03 MJ/kg(ash), 0 kg CO₂/kg(ash)) were summarised to finally obtain 4.23 MJ/kg(ash) and 0.52 kg CO₂/kg(ash) for EE and CF, respectively.

For calculating SUB-RAW index, a conversion was made from kg(ash) to kg(P) assuming 8.45% of P in SSA, resulting EESUB= 50.13 MJ/kg(P) and CFSUB = 6.21 kg CO₂/kg(P). If the temperature conditions were considered, the results would have been slightly higher, but as mentioned in Section 2 any increase of leaching/precipitation temperature regarding wet processes was neglected. In this process, NaOH contributes to the overall emission only for 5.4%. The SUB-RAW index value is 0.06 (Fig. 6) when combining the obtained values with the values set for the reference process (EERAW = 102.32 MJ/kg(P), CFRAW = 4 kg CO₂/kg(P)) (Gilmour, 2013; Hutchins, 1993), suggesting that this process is more suitable with respect to the reference process in terms of environmental sustainability. The positive outcome mainly relies on the different quantity of sulphuric acid used: indeed, in the reference process, sulphuric acid was used for leaching, while in the Lotus Project it was used only for washing the solid residue. Therefore, given that the EE and CF for its production were respectively equal to 7.36 MJ/kg(reagent) and 0.21 kg CO₂/kg(reagent), this difference is justified.

4. Discussions

Fig. 4 shows the EE and CF of the P-extraction methods reported in Table 1. The value corresponding to the reference process (marked with "RAW") for P-extraction from phosphate rocks is also shown. All the data were normalized to 1 kg(P). The EE and CF are mostly enclosed in the range of 0e300 MJ/kg(P) and 0e16 kg CO₂/kg(P) respectively (the limits were set for clarity of visualisation: 7 processes out of 38 were not shown because they had out of range EE and CF values). The reference point approximately determines the region wherein the P-recovery technologies should belong in terms of sustainability evaluation in view of SUB-RAW index elaboration. This "more sustainable area" relatively to the reference technology regarding the environmental sustainability is highlighted by the shaded green background, whereas the technologies outside this area are less sustainable and fall in the red-shaded area (Fig. 4). These two areas are separated by an uncertainty zone (white area in Fig. 4) due to the calculation made for the reference technology, including also the uncertainty reflected by the reagents emission values taken from Ecoinvent database. Among the technologies marked with * in Table 1, Leachphos, Biocon, Lotus Project and ICL Fertilizers technologies were in the "more sustainable area" in the SUB-RAW index chart representation (Fig. 6). ICL Fertilizers' process was similar to the reference process but uses SSA as raw material instead of phosphate rocks. Indeed, these technologies (marked as RAW and 6 in Fig. 4, respectively) have similar EE and CF values because they use similar resources and electrical components. The difference in EE was mainly related to the quantity of the sulphuric acid used (0.6 kg/kg(ash) for the ICL Fertilisers' process and 0.84 kg/kg(ore) for the Rhone-Poulenc process). In addition, differences were the generated steam (0.285 kg/kg(ore)) for the vaporisation and the use of a crushing pre-treatment step needed in the Rhone-Poulenc process and not required for SSA. Fig. 6 shows the final result of the analysis, representing all the SUB-RAW indexes of the

recovery processes of Table 1. The x-axis corresponds to the reference process (SUB-RAW index=0) and all the SUB-RAW index values are between 0.5 and ≈ 1 , a range much smaller than that calculated in other kinds of materials (Bontempi, 2017b). The data points were classified considering three features: (i) availability of input data for each process, (ii) the presence of a purification step in the process (for example, heavy metal depollution), (iii) the material resulting from the process (namely, P₂O₅-based precipitations, H₃PO₄ solution or P₄). More sustainable technologies in comparison to the reference technology (x-axis) have values above the reference line (SUB-raw index>0), while those below the reference line (SUB-RAW index<0 but close to zero (SUB-RAW index=0.17)). In particular, Sephos product is composed of an insoluble aluminium rich precipitate (Nakagawa and Ohta, 2018; Schaum et al., 2007). The process could be improved by adding a further step to obtain a soluble product good in agriculture as already suggested (Nieminen, 2010). However, this step may result in a negative SUB-RAW index, and thus the technology would be less sustainable. Generally, it was observed that parameters such as temperature, resins for cation/anion exchange (e.g. EDTA) or some leaching reagents (e.g. HCl) play a key role in the sustainability of a technology. Therefore, the layout of a process (i.e. electrical and thermal configurations, the ratio between the number of reagents and the amount of entering input material) is mandatory to determine the best condition to reduce environmental impacts. For example, the SUB-RAW of Recophos Inducarb (Langeveld, 2018) and Thermphos (Schipper et al., 2001) processes are 0.47 and 0.24 respectively, and this is explained by the fact they are both thermo-reductive processes. Therefore, the electricity consumed for exploiting inductively thermal heating has a great negative influence, even if P₄ can be obtained. This observation highlights the usefulness of SUB-RAW index, that is a straightforward assessment of the process environmental sustainability and allows to start searching for the critical steps that should be optimized. Moreover, this index can be applied to compare any kind of material or process and have a visual feeling of their relative environmental impacts. In conclusion, ESCAPE approach is able to compare all the available technologies, even if developed only at laboratory scale, offering several advantages, in comparison to limited opportunities proposed by other time-consuming methods, such as LCA. Due to its simplicity and few required parameters, ESCAPE appears to be a new, suitable, and simplified tool, that allows to bring out new technologies for waste, residues, and by-products valorisation, so promoting the circular economy strategy. Moreover, it is important to highlight that the proposed approach is based on a simplified strategy to evaluate the environmental sustainability of a new technology, taking into account only two parameters. Then, it can be considered a preliminary sustainability evaluation step that can allow obtaining an overview of several processes and making the preliminary screening to select only the most promising technologies. After that, a full life cycle evaluation method, such as LCA, can be used to have a full complete study about the environmental sustainability of a technology, involving all the parameters that need to be considered. However, ESCAPE has several advantages: i) it can be applied to compare technologies even if at laboratory-scale, or if the data for the process industrialisation are still not available; ii) due to its simplified approach, it allows to save time and resources, making it suitable for preliminary studies; iii) it allows to bring out new technologies for waste, residues, and by-products valorisation, that cannot be evaluated by LCA approach, promoting technological advance for circular economy strategy implementation; iv) in comparison to more complex approaches, the SUB-RAW index based only on two parameters can be easily understood also by non-specialists, such as legislators and public authorities, v) it can be used to promote, accelerate and support the European policy towards the recycling of materials in order to reach the “zero waste” generation.

5. Conclusions

This paper compares the available SSA P-extraction technologies with the aim to evaluate their sustainability using a novel and simplified approach.

The results show that the choice of the sulphuric acid-based wet process over other type of processes (e.g. wet process using other acids and thermo-reductive dry process), in the context of P-extraction processes, seems to be suitable to be used as a reference process for the calculation of the SUB-RAW index. A relevant

result is related to ICL Fertilisers process, which differs from the reference process only for the input of SSA material and it is shown to be environmentally more sustainable in terms of EE and CF, with its SUB-RAW index equal to 0.14. In view of decreasing the exploitation of phosphate rocks, this result is relevant since a wet chemical leaching process characterises more than 80% of P-extraction processes.

The results also show that many already established wet-based recovery processes (e.g. Biocon, Leachphos, Lotus Project, Sephos) seems to be promising. The Lotus Project process, using wet alkaline-leaching method, is the only one with a positive SUBRAW index (0.06).

Meanwhile, thermo-reductive based dry processes such as Recophos Inducarb and Thermphos (with their SUB-RAW indexes equal to 0.47 and 0.24 respectively) appear to be less sustainable, in comparison to the reference, due to the energy need for the inductively thermal heating. Thus, these technologies seem to be more suitable to produce pure P_4 instead to be used to obtain P (i.e. for fertilisers applications).

Author contributions

Conceptualization: Elza Bontempi, Ario Fahimi Data curation: Elza Bontempi, Ivano Vassura, Ario Fahimi Formal analysis: Ario Fahimi Funding acquisition: This work was supported by DEASPHOR project (Ref: CUP D81118000190002; FCT ERA-MIN/0002/2017) and by FANGHI project, financed by Regione Lombardia, in the frame of the call BANDO Call HUB Ricerca e Innovazione. Investigation: Ario Fahimi Methodology: Elza Bontempi Project Administration: Laura Eleonora Depero Resources Software: Elza Bontempi, Ario Fahimi Supervision: Elza Bontempi Validation: Francesca Ceruti, Laura Cutaia Visualization: Francesca Ceruti, Laura Cutaia Roles/Writing e original draft: Ario Fahimi Writing e review & editing: Elza Bontempi, Laura Eleonora Depero, Bruno Valentim, Stefania Federici, Ario Fahimi

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.

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Acronyms	
P	phosphorus
SSA	sewage sludge ash
PA	phosphoric acid
P ₄	white phosphorus
CF	CO ₂ footprint
EE	embodied energy



Fig. 1. Overview of the main phosphate rocks mining areas (data from (Granta Design, 2019)). The table on the right of the figure shows the % P₂O₅ region by region (data from (Ptáček, 2016))

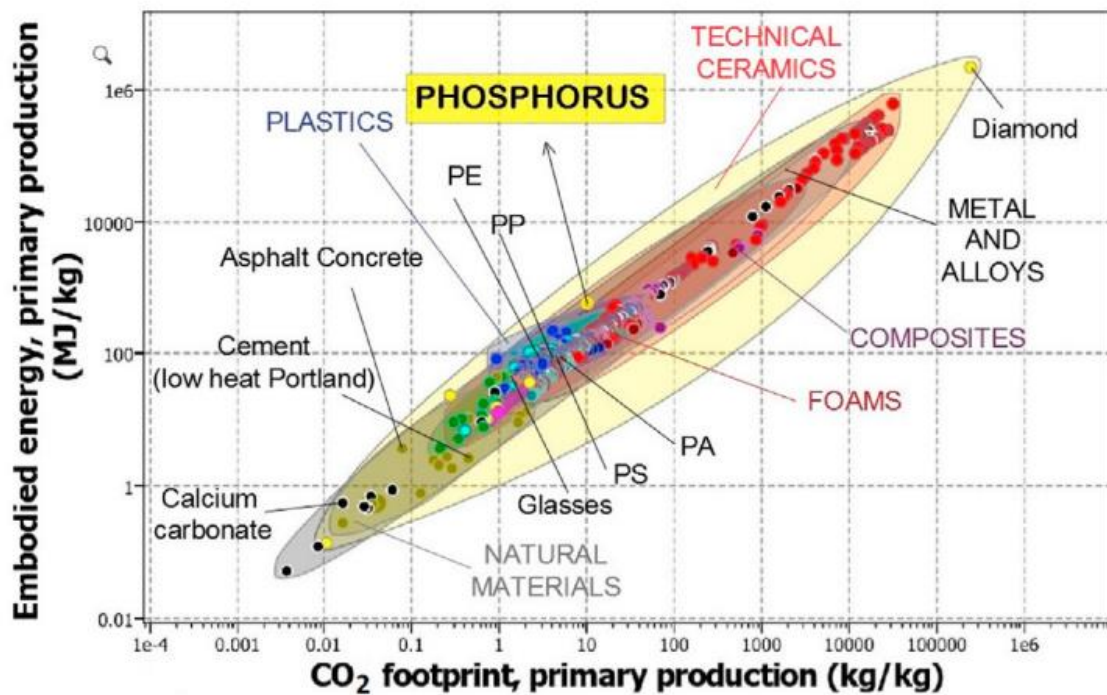


Fig. 2. EE and CF for several materials (data provided by CES Selector, 2019). P is also reported. PE, PP, PS and PA stand respectively for polyethylene, polypropylene, polystyrene and polyamide.

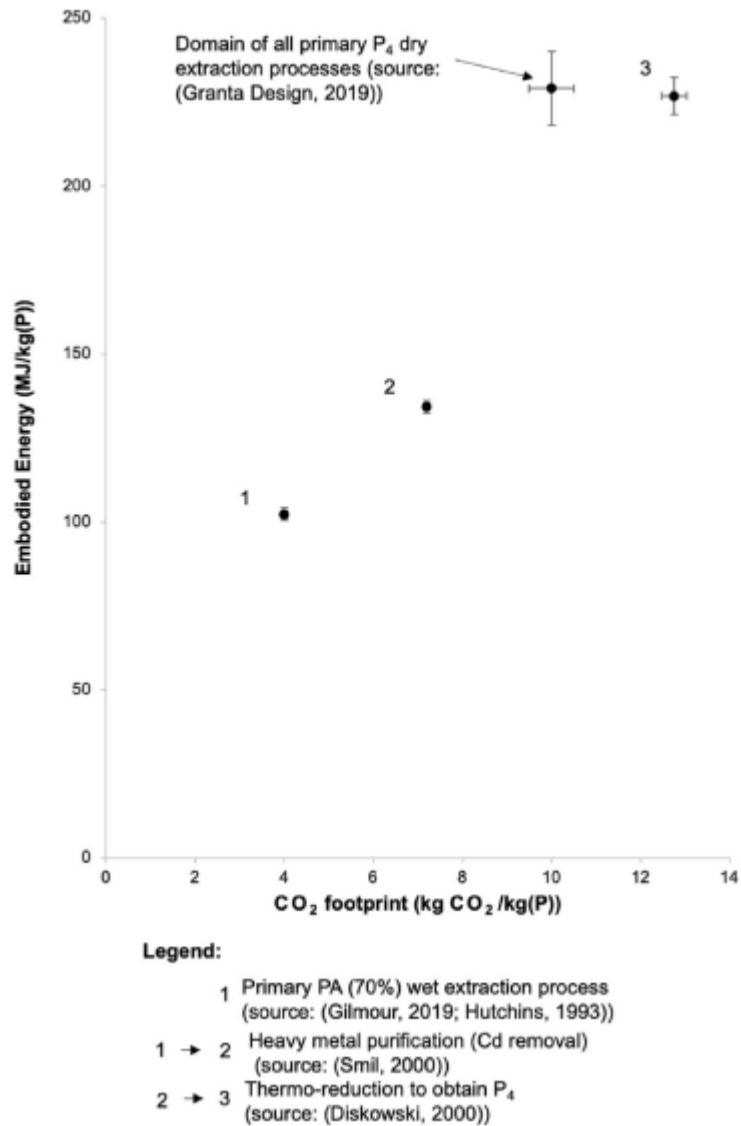


Fig. 3. Environmental emissions comparison between sulphuric acid based P-extraction for phosphate rocks (Gilmour, 2013; Hutchins, 1993) with certified data derived from commercial Ecoinvent v. 2.2 database. The domain enclosing the point marked with “3” represents the mean variation of EE and CF (25%) that corresponds to a decimal unit (± 0.1) variation in a logarithmic scale (SUB-RAW index scale).

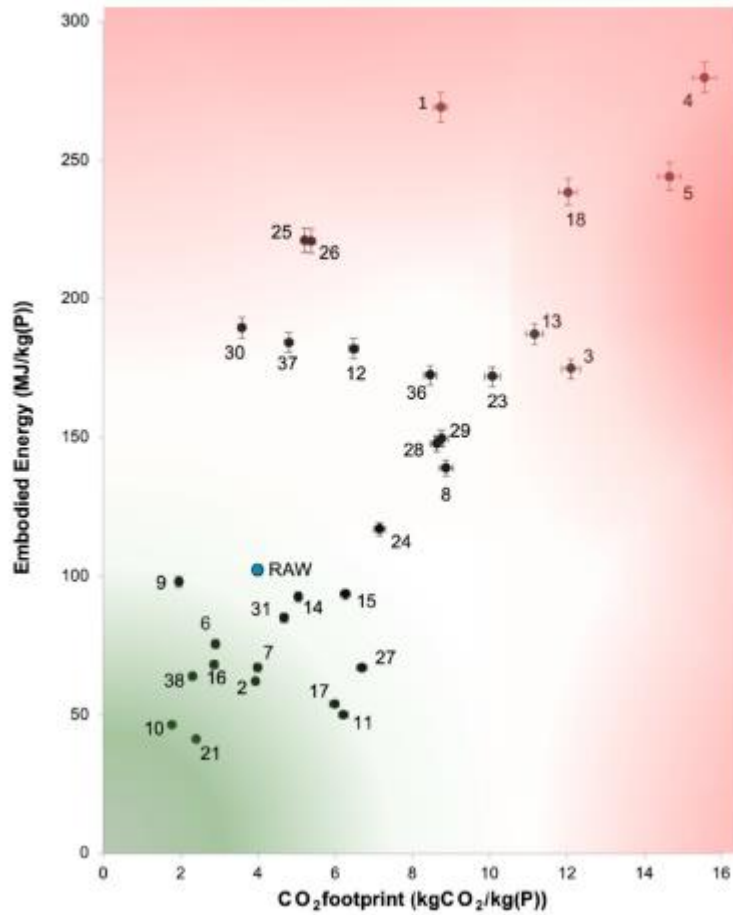


Fig. 4. EE and CF for all the P-recovery technologies listed in Table 1 (all of them indicated on the chart as points): the reference process (Hutchins, 1993) is marked with “RAW”; ICL Fertilizers’ process (Langeveld, 2018), which is the dual sulphuric acid based P-extraction technique with SSA as input material, is marked with “6” (listed as the other processes in Table 1). The colours approximately indicate, the areas in which processes could be more (green) or less (red) sustainable relatively to the reference technology (1) regarding the environmental sustainability calculated via SUB-RAW index. Processes nr. 19, 20, 22, 32, 33, 34, 35 are missing because of their out of range higher emissions. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

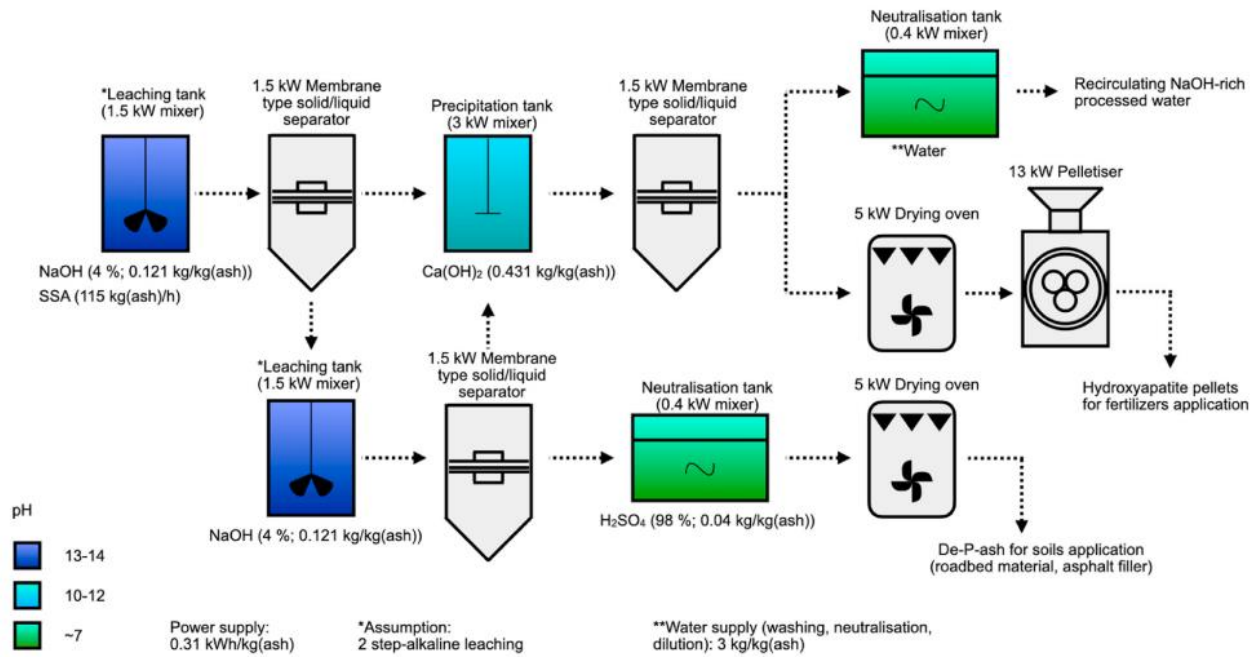


Fig. 5. Simplified flowsheet scheme for the case of Lotus Project (Gifu City, Japan) reported and described in (Nakagawa and Ohta, 2018): input reagent mass flows all referred to the mass of starting waste material.

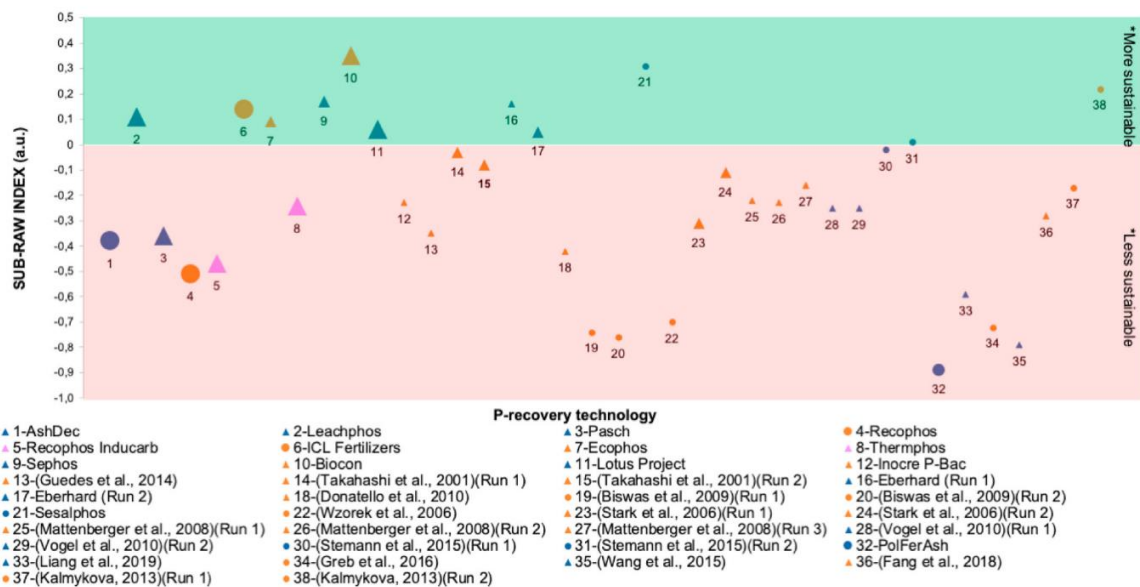


Fig. 6. SUB-RAW index chart representation: the x-axis represents the reference process (Hutchins, 1993); the green shaded area includes the technologies which result more sustainable (SUB-RAW index > 0) and the red shaded area includes the technologies which result less sustainable (SUB-RAW index < 0)

Table 1 Overview of P-recovery technologies from SSA.

Overview of P-recovery technologies from SSA.

Nr.	Technology	Country initiative	Scale	Approach	Reference
1	AshDec ^a	Germany/Switzerland	Pilot	Thermo-chemical	Hermann and Schaaf (2018)
2	LeachPhos ^a	Switzerland	Pilot	Wet chemical leaching	Morf (2012)
3	Pasch ^a	Germany	Laboratory	Wet chemical leaching	Montag et al. (2009)
4	Recophos ^a	Germany	Full	Wet chemical leaching	Montag et al. (2015)
5	Recophos Inducarb ^a	Germany	Pilot	Thermo-reductive	Langeveld (2018)
6	ICL Fertilizers ^a	The Netherlands	Full	Wet chemical leaching	Ten Wolde (2013)
7	EcoPhos ^b	France	Full	Wet chemical leaching	De Ruyter (2014)
8	ThermPhos ^a	Kazakhstan	Industrial	Thermo-electric	Schipper et al. (2001)
9	Sephos ^b	Germany	Laboratory	Wet chemical leaching	Schaum et al. (2007)
10	BioCon ^a	Sweden	Laboratory	Wet chemical leaching	Levlin (2007)
11	LotusProject ^a	Japan	Full	Wet chemical leaching	Nakagawa and Ohta (2018)
12	Inocre P-Bac ^c	Japan	Pilot	Bio-leaching	Chi et al. (2006)
13	ED cell ^c	Denmark/Portugal	Laboratory	Electro-chemical leaching	Guedes et al. (2014)
14, 15	Takahashi et al. (2 runs) ^b	Japan	Laboratory	Wet chemical leaching	Takahashi et al. (2001)
16, 17	Eberhard (2 runs) ^b	Switzerland	Laboratory	Wet chemical leaching	Franz (2008)
18	Donatello et al. ^c	United Kingdom	Pilot	Wet chemical leaching	Donatello et al. (2010)
19, 20	Biswas et al. (2 runs) ^c	Japan	Laboratory	Wet chemical leaching	Biswas et al. (2009)
21	SesalPhos ^c	Germany	Laboratory	Wet chemical leaching	Petzet et al. (2011)
22	Wzorek et al. ^c	Czech Republic	Laboratory	Thermo-chemical	Wzorek et al. (2006)
23, 24	Stark et al. (2 runs) ^b	Sweden	Laboratory	Wet chemical leaching	Stark et al. (2006)
25, 26, 27	Mattenberger et al. (3 runs) ^c	Austria	Pilot	Thermo-chemical	Mattenberger et al. (2008)
28, 29	Vogel et al. (2 runs) ^c	Germany	Laboratory	Thermo-chemical	Vogel et al. (2010)
30, 31	Stemann et al. (2 runs) ^c	Germany	Laboratory	Thermo-chemical	Stemann et al. (2015)
32	PolFerAsh ^b	Poland	Laboratory	Wet chemical leaching	Gorazda et al. (2017)
33	Liang et al. ^c	China	Laboratory	Wet chemical leaching	Liang et al. (2019)
34	Greb et al. ^c	Germany	Laboratory	Wet chemical leaching/Thermo-chemical	Greb et al. (2016)
35	Wang et al. ^c	China	Laboratory	Wet chemical leaching	Wang et al. (2018)
36	Fang et al. ^c	China	Laboratory	Wet chemical leaching	Fang et al. (2018)
37, 38	Kalmykova (2 runs) ^c	Sweden	Laboratory	Wet chemical leaching	Kalmykova and Karlfeldt Fedje (2013)

^a All the data for the process evaluation are provided in an exhaustive way.^b Partial availability of all necessary data related to reagent input mass flow, EE and CF of reagents or power consumption of electrical components are lacking.^c The majority of required data are lacking. Further information about the assumptions are indicated in the supporting information (S1).