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Closing the nutrient cycle by using organo-mineral fertilizers based on secondary raw materials

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Abstract
The increasing in nutrient flow through agri-food system caused by population growth and rapid urbanization has contributed to disruption of natural biochemical cycles. Ineffective use of nitrogen and phosphorus has resulted in high nutrient losses to the environment and consequently leaded to eutrophication process, soil and air pollution. And the same time, the huge problem with management of nutrient-rich waste is existed. In accordance with new European policy, which is focused on optimizing resource and energy use, waste should be recycled. The aim of this study was to characterize the fertilizer potential of sewage sludge, manure and by-products after its treatment in the context of its use as secondary raw materials in organo-mineral fertilizer production. Taking into account nutrient content, pollution load and availability of collection and treatment processes the technology of fertilizer production based on sewage sludge and poultry litter ash is proposed. The characterization of fertilizers for rape, flax and sunflower crops is presented.

Keywords: sewage sludge, manure, nutrient recycling, organo-mineral fertilizer, circular economy

1. Introduction

Global population is projected to increase from 7.6 to 9.8 billion by 2050 [1]. According to FAO data, average calorie intake has increased from about 2,000 kcal/capita/day in the 1960s to more than 2,800 kcal/capita/day today. Population growth and changes in dietary patterns including higher meat and dairy consumption is a great challenge for agriculture. It is estimated, that demand for crop will increased by 70-110% by 2050 [2]. Meeting the need for agriculture products will be connected with increasing fertilizers use, which are the primary source of nutrients.

Mineral fertilizer market is very consolidated especially in the case of phosphorus and potassium fertilizers. Phosphate rock resources, 81% of which is used for fertilizer production [3] are concentrated only in few countries. Morocco, China, Algeria, Syria and South Africa controlled 84% of phosphate rock reserves [4]. Taking into account an uncertainty surrounding the level of global phosphate deposits and a decreasing of phosphate ore quality there is a concern related to steady supplies for courtiers-importers [5]. Phosphorus fertilizer industry in Europe is dependent on imported raw materials [6]. Therefore, the European Commission has added phosphate ore to the list of critical raw materials as a resource with high economic importance. It is assumed, that phosphorus inclusion in the critical raw materials list will promote actions towards improving phosphorus use efficiency in the near future [7].

Information on the potassium deficit is limited in scientific literature. With current consumption rates potassium ore are expected to last 330 years. Nevertheless, the uneven distribution of potash reserves is observed. The largest potassium ore deposits are located on the territory of Canada, Belarus and Russia. The share of these countries in global reserves accounts for 58% [8]. The price for potash is strongly controlled by two main exporters – Canada and Russia. In these circumstances there are only few countries, which are self-sufficient in potassium using mineral fertilizers [9].

The manufacture of nitrogen fertilizers is based on Haber-Bosch process, which uses fossil fuels. It is estimated, that nitrogen fertilizer production is responsible for the consumption of 1-2% of global energy [10]. Therefore, sustainability of nitrogen fertilizer manufacture is strongly dependent on energy prices, which was observed in 2008 when prices of fertilizers had increased because of increasing prices of oil. An important issue related to nitrogen fertilizer production is contribution in significant amount of greenhouse gas (GHG) emissions. Global estimated amount of CO_2-eq generated by nitrogen fertilizer industry accounts 410 million tons per year [11].

There is no doubt that mineral fertilizer use has allowed a tremendous increase in world food production. However, the establishment of agriculture have led to disruption of natural biogeochemical cycles of nutrients, which has adversely effect on the environment and human health. Only 20% of nitrogen entering in agricultural system is converted to final product for consumption. In case of phosphorus nutrient use efficiency is 30%. Such low efficiency of production chains contributes to nutrient leakage to water and air [12]. Dissipation of nitrogen and phosphorus makes a serious threat for aqua-system causing the eutrophication process. Uncontrolled nutrient flows in agriculture strongly impact climate. In general the share of greenhouse gases emitted from agricultural sector in European Union is 10%. The nitrogen surplus from manure and mineral fertilizers can lead to soil acidification, acid rain, formation of particulate matter which results in reducing of soil fertility and microbial activity, water pollution, damaging of yield and forests and causing respiratory disease in humans [13]. It is estimated, that fertilizer value of nitrogen losses from agriculture into the environment is around € 20 billion per year [12].

2. Circular economy concept

High nutrient inputs in agro-food system result in generation of huge amounts of food chain waste, human and animal excreta. The disposal of this waste is often a great challenge for communities. However, it seems to be a chance for creating sustainable nutrient management by closing nutrient loop through safety recycling and recovery of nutrients from this waste streams. Such approach is a solution addressing two concerns: management of waste containing beneficial nutrients on the one hand and reduction reliance on imported natural resources using for mineral fertilizer production on the other.

Circularity of nutrients in production chains is a key priority in new business model, which is an alternative for traditional economy with “take-make-use-dispose” pattern. Circular economy is based on an assumption that the value of products, materials and resources should be maintained as long as possible to ultimately minimize waste generation. It suggests to use waste and by-products from one stage of the production to another stage giving them a new market value. Circular economy makes a possibility to decrease a gap in natural resource requirements and supply shortages though the closing the loop of material flow [14-15].

The circular economy goal is promoting innovative and more efficient way of production/distribution and consumption. Shifting of this business model will bring economic and environmental benefits. It is estimated, that efficient use of resource is able to decrease materials inputs in European Union by 17-24% by 2030. Prevention of waste generation can save 600 billion euro in business in EU, and in addition reduce greenhouse gas emissions by 2-4% [16].

The core principle of circular economy is reduce, reuse and recycle [17]. This concept emphasizes total value recovery [18]. Therefore, closing the loop of nutrient flows should take into account the potential of organic matter which is in waste streams. This issue is usually overlook in nutrient management. And organic matter plays a key role in soil fertility by affecting its physical, chemical and biological properties. Intensive agricultural practices have led to significant decrease of organic matter content in soil. According to the European Soil Database (ESB), about 45% of European soils are characterized by low and very low content of organic carbon [19]. Due to this, possibility of organic matter recycling together with nutrients is a matter of high importance.

Circular economy offers transformation of nutrient-rich waste into secondary raw materials and encourages their use in fertilizer production on a large scale. It allows to improve efficiency of natural resource use, which is crucial in case of finite phosphorus reserves, reduce costs related to waste management and decrease environmental pollution caused by nutrient leakage and processes associated with mineral fertilizer production (Figure 1).

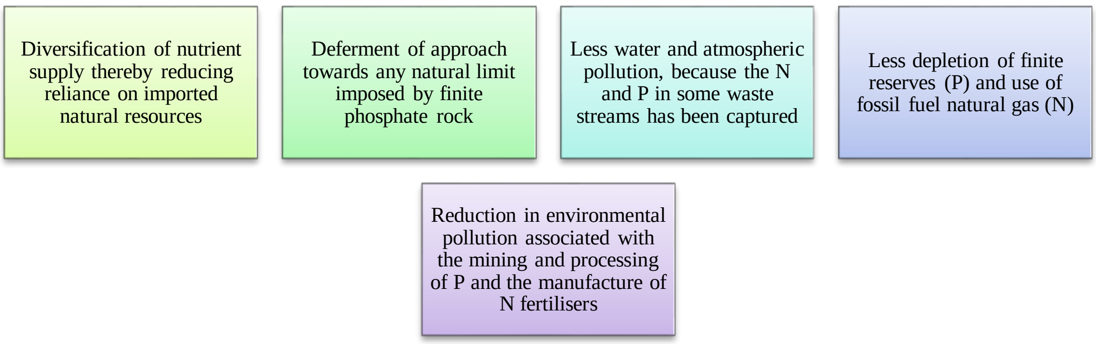

Figure 1. Benefits of nutrient recycling and recovery [12]

Among the different waste streams sewage sludge and manure gain the most interest for nutrient recycling and recovery because of their abundance, ubiquitous presence and concentration of nutrients.

3. Characterization and disposal methods of main waste streams suitable for nutrient recycling and recovery

Sewage sludge

Sewage sludge is an unavoidable waste generated during wastewater treatment. Due to the implementation of the Urban Waste Water Treatment Directive in European Union the amount of sewage sludge is increasing. More than 9 million ton of sewage sludge expressed as dry solids was generated in EU in 2015 [20].

The appropriate management of sewage sludge is a great challenge because of its specific composition and costs of disposal, which usually accounts 50% of wastewater treatment plant operating costs [21].

Sewage sludge is semi-solid organic waste, rich in primary nutrients, macro- and microelements. Typically, sewage sludge Typically, sewage sludge contains 1-5% of P in dry mass and up to 8% if biological phosphorus removal was applied [22]. applied [22]. Nitrogen concentration varies from 0.1-18% (N) and usually reaches 3.3% [23]. High concentration of of nutrients and organic matter makes sewage sludge an attractive material for fertilization. Using sewage sludge for for agricultural purposes is well-known and widespread practice considered as an essential element of sustainable nutrient nutrient management. More than 50% of sewage sludge is applied to soil in European Union. It is preferred option of sewage of sewage sludge disposal in United Kingdom, Ireland and Spain, where more than 70% of sewage sludge is used in in agriculture, as well as in Bulgaria, Denmark and France, where share of sewage sludge applied to soil accounts approximately 50% (

Figure 2). Many studies have shown a positive effect of sewage sludge application on soil and crop yields, including improvement of soil structure and porosity, increasing ability to exchange cations and enzymatic activity of microorganisms, increasing of nutrient and organic matter content in soil and high growth of plant biomass [24-29].

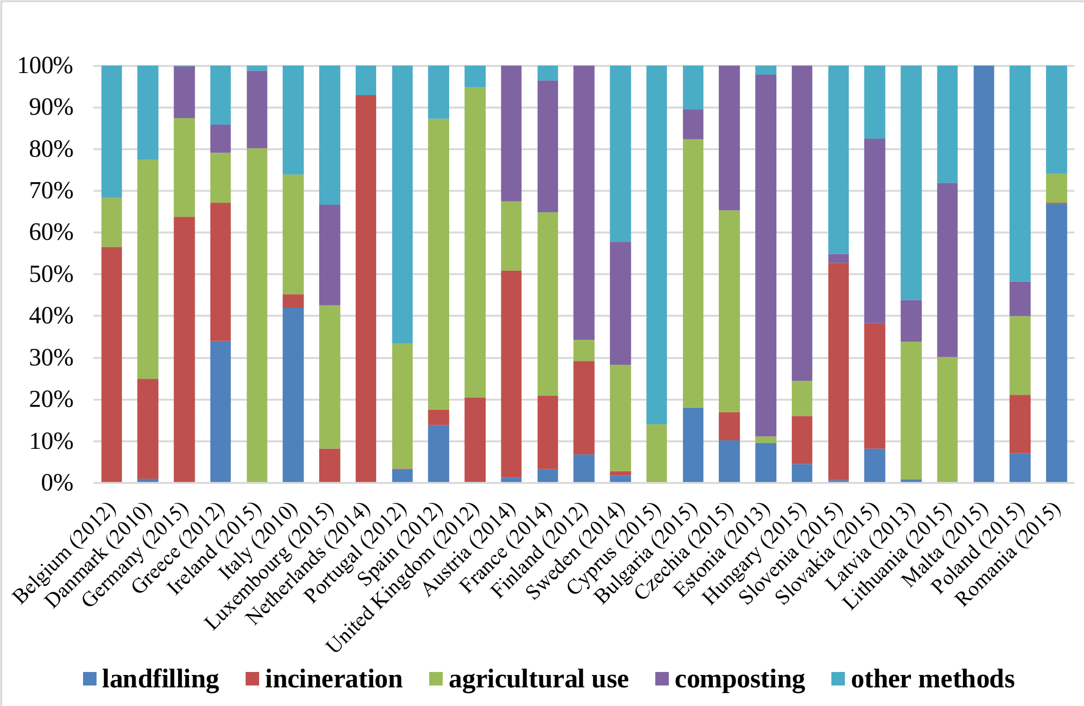

Figure 2. Methods of sewage sludge disposal in European Union countries [30]

However, sewage sludge use in agriculture can be limited because of possible presence of heavy metals, toxic organic compounds and pathogenic microorganisms. The heavy metal content in sewage sludge is diverse on EU countries and even in the same country (Table 1). Studies have shown, that high heavy metal concentration is usually found in sewage sludge originated from industrialized cities. And sewage sludge generated in small WWTPs do not pose threat for environment in respect of heavy metal content [31-32].

Table 1. Heavy metal content in sewage sludge in European Union countries [31-37]

Cd	Cu	Pb	Zn	Cr	Ni	Hg	Country	n

mg/kg DM								
0.7	269	48.3	505	-	25.1	0.7	UK	28
<0.3-1.0	140.8-155.8	<5.6	581.1-757.2	<5.6	22.6	<1.3	PT	2
5.87	143.7-380.4	23.6-167.8	770.3-1,613.6	26.4-114	13.6-140.5	0.5-1.2	PL	2
0.67	174.88	42.94	-	46.21	32.42	-	SP	1
0.35-19.4	97.9-339.4	5-124	174.7-1,437.5	16.7-811.3	7.8-70	0.1-1.95	LT	7
0.8-7.3	51-198	12-102	810-1,880	12-355	8.8-64	-	GR	3
0.331-1.357	455.2-727.2	18.7-70.69	286.3-1,260.8	39.58-57.16	21.55-34.09	0.58-1.73	IT	3

n – number of samples

There are concerns related to persistent organic pollutants (POPs), which can be occurred in sewage sludge due to a lack of knowledge related to its transformation pathways. During wastewater treatment processes the most of easily degradable contaminants are removed. Nevertheless, some groups of POPs with lipophilic properties can adsorb on sludge particles and get into the environment when sewage sludge is applied to soil [38]. Among the most dangerous organic pollutants presented in sewage sludge PAH (polycyclic aromatic hydrocarbons), PCB (polychlorinated biphenyls), PCDD/F (polychlorinated dibenzo-p-dioxins and dibenzo-furans), NPE (nonylphenol and nonylphenol ethoxylates), PFC (perfluorinated chemicals) and DEHP (di-(2-ethylhexyl)phthalates) can be mentioned. Much attention have to be paid on occurrence of pharmaceuticals and personal care products in wastewater systems due do its increased use and persistent in the environment. The range concentration of organic contaminants found in sewage sludge from different literature sources are collected in [38], [39].

Sewage sludge can contain pathogenic microorganisms such as bacteria, viruses, protozoa and other parasitic worms, which pose an epidemiological threat and may cause various allergies, act toxic or immunotoxic to humans and ecosystem. The amount of pathogenic microorganisms can be significantly reduce or completely eliminate by using appropriate sewage sludge treatment processes (digestion, composting, liming, drying, pasteurization etc.) [40]. Nonetheless, these methods are not effective in case of viruses. Such viruses as *Herpesvirus*, *Papillomavirus*, *Adenovirus*, *Bocavirus*, *Klassevirus*, *Coronavirus* and *Rotavirus* are still identified in sewage sludge after treatment processes [41].

In order to prevent negative impact of sewage sludge application on the environment, animals and people its agricultural use is regulated by a Council Directive 86/278/EWG [42]. According to the directive sewage sludge intended for soil applications should be appropriate treated and fulfilled the requirements in regards to heavy metals content. The member states of EU have transposed the restrictions related to sewage sludge quality, which can be used in agricultural purposes to their national legislations. Such countries as Austria, Belgium, Czech Republic, Finland, Germany, Netherlands, Slovenia and Sweden have set up more stricter limits for heavy metals content in sewage sludge. Additionally, Austria, Denmark, France, Germany and Sweden have adopted permissible values for toxic organic compounds in sewage sludge. The content of pathogenic organisms such as *Salmonella, Enterobacteria, Escherichia coli, Enterovirus, helminths* are regulated in sewage sludge intended to soil in France, Denmark, Luxembourg, Poland, Finland and Italy [43].

The identification of key sources of pollutants as well as national and European environmental regulations play a crucial role in reduction of toxic compounds release to the environment. Study conducted by Zennegg during 20 years showed, that a ban or a restriction on the use of PCB and PCDD/F have resulted in concentration decrease of these substances in sewage sludge at 69-83% [44]. Environmental policy undertaken in Sweden related to improving sewage sludge quality has contributed to reduction of heavy metals from 1970 at average 85% [45]. It was also reported significant decrease of heavy metal concentration in sewage sludge in Germany in years 1977-2006 (95.4% for Cd, 94.2% for Cr, 87.7% for Hg) [46].

Agricultural use of sewage sludge is the most attractive option of its disposal mainly due to the low costs and possibility for nutrients and organic matter recycling. However, there is some technical problems associated with lack of surface, where sewage sludge can be applied and the fact, that sewage sludge are generated throughout the year, but can be used 2-3 times a year [47].

Bearing in mind the restrictions related to organic recycling of sewage sludge and possible occurred pollutant load, incineration seems to be optimal solution for sewage sludge management. Thermal treatment of sewage sludge allows for potential energy recovery and significant reduction of mass and volume (up to 80-90%) together with decomposition of harmful substances and pathogenic microorganisms [48], [49]. However, it should be emphasised, that investment and operating costs of sewage sludge incinerator plants are very high. From this point of view, sewage sludge combustion are profitable only for large agglomerations [50]. Additionally, the disadvantage of sewage sludge incineration is that it is not a zero-waste method. The ash remaining after combustion process characterized by high heavy metal content therefore should be disposed in environmentally safe way [51]. There are reports about possible formation of PCDD/Fs during sewage sludge incineration, which can be absorbed on the ash surface increasing its toxicity [52]. On the other hand, sewage sludge ash is rich in phosphorus (10.0-25.7% P_2O_5) and seems to be a promising material for fertilizer production [53, 54].

Eurostat data shows, that near 26% of sewage sludge is incinerated in Europe. In such countries as the Netherlands, Netherlands, Germany, Belgium, Austria and Slovenia over 50% of sewage sludge is directed to thermal utilization. In utilization. In contrast to this, Ireland, Cyprus, Bulgaria, Latvia, Croatia, Lithuania and Malta do not use this sewage sludge sewage sludge disposal method (

Figure 2) [30].

Currently, the European policy is aimed at reducing the amount of sewage sludge which is landfilled mainly due to the economic and environmental issues and promotes the maximum recovery of material and energy contained in sludge. However, landfilling and so-called other methods for sewage sludge disposal (included temporary or long storage at WWTPs, landfill cover, reuse in green area and forestry, etc.) still play an important role in Romania, Malta, Italy, Greece, Hungary, Latvia, Hungary, etc. [30, 55].

Manure

The livestock population in Europe is one of the world's largest, accounts 148 millions of pigs, 88 millions of cattle and 1,428 millions of chicken. This animals generate annually approximately 1,400 million tons of manure [56]. The primary factors effect manure composition are animal type and age, feeding practices and production volume of milk/meat/eggs.

Manure is regarded an attractive organic fertilizer with high nutrient content (Table 2). Nitrogen and phosphorus in manure are presented in organic and inorganic forms. Inorganic nitrogen is generally found in ammonia form and urea. The proportion of organic and inorganic nitrogen forms depends on animal type and determines potential of manure as fertilizer and potential environmental losses [12]. Despite high nitrogen and phosphorus content in manure its ratio is lower than required for plants, which can lead to phosphorus accumulation in soil. Potassium in manure is presented in soluble form and can be easy absorbed by plants from soil solution [57].

Table 2. Nutrient content of different types of manure [58]

	Dry matter (%)	Organic matter (% of DM)	Potassium (% K of DM)	Nitrogen (% N of DM)	Phosphorus (% P of DM)
Liquid cattle manure	3	57	29.4	12.2	1.0
Solid cattle manure	22	64	2.1	2.4	0.7
Liquid pig manure	2	n.a.	9.1	17.1	3.2
Solid pig manure	24	80	2	3.2	1.4
Solid poultry manure	57	74	2.1	4.4	1.4

The main disposal method of manure in European Union is agricultural use. It is estimated, that more than 90% of manure is applied to soil by spreading collected manure or grazing activities [59]. However, in regions with intensive livestock production manure management is a big challenge. For example, in Finland 60% of phosphorus and 33% of nitrogen in agriculture comes from manure. The calculations show, that in some regions the total amount of phosphorus from generated manure that exceeds the crop fertilizing requirements reaches 20% of annual manure phosphorus quantity [60]. In Norway the demand for phosphorus is estimated to be 5,800 tons per year when taking into account the amount of bioavailable phosphorus in soil. And phosphorus applied every year to Norwegian soils as manure is 12,000 tons [61]. A common practice for manure disposal is a transport of manure to regions where there is a deficit of nutrients in the soil, which is associated with substantial costs due to high water content in manure. The agricultural use of manure is also limited by the Nitrates Directive, which establishes maximum load for nitrogen equal 170 kg N/ha and national legislation in some European countries, which regulates maximum phosphorus application rates [62].

Due to animal by-product regulation manure belongs to category 2 and there is no requirement for treatment of manure, treatment of manure, which can be applied to soil [63]. Only 7.8% of manure in European Union is processed [59]. Application of unprocessed manure to soil involves the risk with accumulation of pollutants including heavy metals, metals, antibiotics and pathogens. The main source of heavy metals in manure is feed additives, which are used in order to in order to promote animal growth [64]. The concentration of heavy metals in manure is varied between different animal animal types. Pig manure was found to be the most contaminated by heavy metals, especially of Cu and Zn, which is related is related to the specificity of their diet (

Table 3).

Table 3. The range of heavy metal content in different types of manure [65]

	n	Cu	Zn	As	Cr	Cd	Pb
		mg/kg DM					
Cattle manure	48	10.28-112.90	16.97-377.17	0.46-19.44	nd*-3.60	nd-10.49	0.53-5.43
Chicken manure	34	1.53-487.43	15.37-1,063.32	0.55-10.42	nd-2,402.95	nd-37.99	nd-22.1
Pig manure	36	77.62-1,521.43	63.37-1,622.81	1.00-33.48	nd-43.45	nd-203.40	nd-5.08

nd – non-detectable
n – number of farms

The global consumption use of antibiotics in livestock production is estimated 63,151±1,560 tons in 2010 and will increase to 105,596 ± 3,605 tons in 2030 [66]. Majority of antibiotics is extracted with urine and feces (75-90% of the ingested doses) and when applied to soil can be uptaked by crops and contribute to elevated antibiotic resistance and allergies in animals and humans [67], [68]. The most-used groups of antibiotics in animal husbandry in European countries are tetracyclines (32.8%), penicillins (25.0%) and sulfonamides (11.8%) [69]. Since 2006 the using of antibiotics for growth promotion of animals is forbidden in Europe. However, due to the increasing numbers of farms the amount of used antibiotics did not decreased [70]. There are reports about occurrence of bacteria (*Salmonella*, *Escherichia coli*, *Yersinia*, *Campylobacter*, *Clostridium perfringens*), protozoa (*Giardia*, *Cryptosporidium)*, helminths *(Ascaris lumbricoides*, *A. suum)* and viruses (*Norovirus*, *Rotavirus*, *Astrovirus*, *Teschoviruse*) in manure, some of which are particularly persistent and can cause human diseases [62], [71] .

Storage, handling and spreading of manure is associated with serious environmental problems. Manure is a significant source of greenhouse gases (GHG) and ammonia. It is estimated, that livestock farming in Europe contributes to 80% of total ammonia and 10-17% of total GHG emissions. Additionally, the excess of nutrients from over application of manure contributes to eutrofication process, soil acidification and pollution of surface, groundwater and loss of biodiversity [72], [73]. Pollution costs related to manure management in Europe is estimated in 12,300 million € annualy (Figure 3). Therefore, from environmental and economic point of view, it is very important to reduce nutrient leakage and gaseous emissions associated with manure. A number of treatment technologies capable to minimize nutrient losses and at the same time increase fertilizer potential of manure are available in the market. However, the implementation of this technologies in practice within member states is very low. National policies as well as awareness and perceptions of pivotal stakeholders have the main influence on increasing of manure processing extent [72].

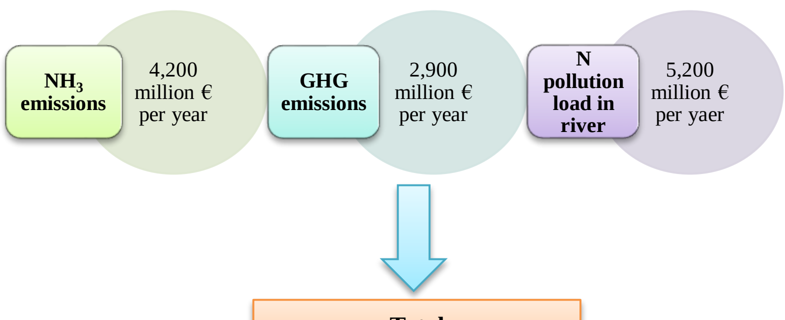

Figure 3. Estimated environmental impact costs associated with manure management [56]

The most common treatment technology used in European countries is anaerobic digestion and liquid/solid separation. Anaerobic digestion is applied for 6.4% of manure generated in Europe. In case of liquid phase after manure separation 0.7% is further processed by evaporation, filtration or though biological treatment (nitrification-denitrification processes) [59]. Solid phase of manure can undergo composting, drying and pelletizing or combustion. Thermal processes are mainly concerns poultry manure due to higher dry matter content. Fertilizer industry shows interest in ashes after manure combustion because of significant phosphorus and potassium content [12]. About 1,500 tons of poultry litter is currently incinerated in Europe by such commercial companies as BMC Moerdijk in Netherlands, Fibropho in United Kingdom, BHSL in Ireland and in some Scandinavian Member States, which allowed to recovery of approximately 0.03 tons of phosphorus annually [74].

4. Nutrient recycling and recovery from sewage sludge and manure

The increasing demand of nutrients in agro-food systems from the one hand and generating huge amounts of waste, unappropriated management of which has negative impact on environment and economy security, on the other hand has contributed to development of technologies for nutrient recycling and recovery. The main challenges facing such technologies are high dilution of waste streams, multiple sources, composition variation, concerns related to possible pollutant occurrence and economic feasibility. Hence, there are three main tasks, which should be undertaken in order to improve nutrient management towards circular economy:

i) improving waste collection and nutrient accumulation by using appropriate treatment method in order to increase the total amount of recycled and recovered nutrients;
ii) identification of key pollutant sources and taking technical and regulatory actions for reducing their emission in order to obtain safe product;
iii) using methods and techniques allowed to increase the fertilizer equivalent of recycled/recovered product, which are stable, easy for transport, storage and application.

The estimated nutrient content of municipal wastewater and manure generated annually in European Union is presented in Table 4. The amount of nutrients in this waste streams is impressive when compared with mineral fertilizer consumed in European Union, which accounted 11.1 million tons of N, 1.1 million tons of P and 2.4 million tons for K fertilizers in 2016.

Table 4. Nutrient content in sewage and manure generated annually in European Union [58], [75].

	Nitrogen (million tons)	Phosphorus (million tons)	Potassium (million tons)
Manure	9.3	2.5	8.0
Sewage	2-3.1	0.5	n.d.*
Mineral fertilizer consumption in European Union in 2016	11.1	1.1	2.4

n.d. – no data available

Unfortunately, not all of these quantities are recoverable, which results from specifics of treatment processes in case of sewage or lack of such processes in case of manure. It is estimated, that in wastewater systems 0.074 million tons of P is lost as effluent, 0.059 million tons is in untreated and uncollected wastewater, 0.227 million tons goes to communal sewage sludge and 0.174 million tons is in sewage sludge, which is used in agricultural purposes [58]. During sewage treatment between a third and a half of the nitrogen is lost during nitrification/denitrification processes [12]. In case of manure the estimations have shown, that due to losses only approximately 52% of nitrogen can be potentially recycled [76].

Currently, recovery technologies are focused mainly on phosphorus due to the scarcity of this resource and nitrogen recovery often has lower priority. A wide spectrum of phosphorus recovery techniques has been developed and implemented mostly in WWTPs and also as an element of manure treatment. Most of them are based on crystallization/precipitation processes and acid leaching or thermochemical methods to produce struvite, hydroxyapatite, Ca- and Mg-phosphates or elemental phosphorus and phosphoric acid, which can be used in fertilizer industry. Phosphorus recovery is possible to achieved from urine after separation, secondary treated effluent, digester supernatant, sewage sludge and sewage sludge ash, slurry after liquid/solid separation, digested manure and biochar [77-80]. Nitrogen recovery usually takes place through struvite precipitation. However due to the fact, that nitrogen in sewage and manure is presented in excess of the molar ratio N:P in struvite (1:1) large quantity of it is remained in effluent [81]. Another approaches for nitrogen recovery are air stripping of ammonia and methods based on ion exchange and adsorption and membrane separation [82, 83]. There is also possibility for potassium recovery together with phosphorus in form of K-struvite from urine [84] or manure [85].

Improving and implementation of recovery technologies is an important step towards circular economy. Nevertheless, closing the nutrient cycle requires total value recovery from waste streams at local, national and European scale not only focus on one or two elements [18].

Fertilizer production from sewage sludge and poultry litter ash

An approach, that offers to reach 100% of nutrient use is fertilizer production based on secondary raw materials. An example of such fertilizers are those produced from sewage sludge. Sewage sludge is a source of organic matter and nutrients in the soil. Nevertheless, sewage sludge can contain heavy metals, POPs and pathogens as was mentioned above. Therefore, the quality of sewage sludge will determine the possibility of using sewage sludge as a material in fertilizer production [86]. Transformation of sewage sludge into organic or organo-mineral fertilizers is not a new idea. Available technologies are generally included treatment process with acid or alkali agents in order to disinfect and stabilize sewage sludge and in some cases adding mineral fertilizers to increase nutrient content of final product. Usually the obtained fertilizers have low nutrient content in comparison with mineral fertilizers and can be classified rather as soil amendments than self-efficient fertilizers [87].

Technologies of sewage sludge based fertilizer production open the opportunity of transformation waste into safe and valuable product. But, attention should be also paid on the right ratio of nutrients depending on the plant's nutritional requirements. This helps to reduce nutrient losses to the soil and surface waters, which is important from environmental point of view.

We proposed technology of balanced organo-mineral fertilizer production characterized by high nutrient content dedicated for rape, flax and sunflower crops based on sewage sludge and poultry litter ash. The technology gives a possibility for management of sewage sludge and poultry litter ash and provides nutrient and organic matter recycling in line with circular economy.

5. Materials and methods

Analytical procedures

The chemical composition of sewage sludge and poultry litter ash were determined with the use of Atomic Absorption Spectroscopy (AAnalyst 300 Perkin Elmer) and ICP-OES (Plasm 40 Perkin Elmer) after digestion in H_2SO_4 and in aqua regia in the case of Ca and Pb determination. Digestion of fertilizers were carried out in aqua regia. The total phosphorus content, as well as its bioavailability was determined by spectrophotometric method according to polish standard (PN-88/C-87015). Determination of nitrogen content was done by using 2400 CHN Elemental Analyser Perkin Elmer. The organic mass content of dried sewage sludge was determined by calcination at 550° C. The analysis was performed in duplicate. Measurements of compressive strength of sewage sludge based fertilizer granules were made by using the ERWEKA TBH 200D apparatus. Each time 20-30 granules were used for the measurement. 30 granules with diameter, which was the main fraction in the test sample, were subjected to testing each time.

Characterization of secondary raw materials

Sewage sludge was collected from WWTP from Żywiec, Malopolska region, Poland. Sewage goes mechanical and biological treatment. Resulting sewage sludge are anaerobically digested and dried on medium temperature belt drier (60-130° C) to water content 5-15%. The main way of sludge management is forwarding to an external company for the production of alternative fuel used in cement plants. The poultry litter ash used in experiment is originated from industrial incinerator plant. Poultry litter was incinerated in rotary kiln at 650-850° C. The chemical composition of secondary raw materials used for fertilizer production is presented in Table 5.

Table 5. Composition of secondary raw materials used for fertilizer production

	OM*	**N**	**P_2O_5**	**K_2O**	**Ca**	**Mg**	**Ni**	**Cr**	**Pb**	**Cd**	**Hg**
	% DM						mg/kg DM				
Digested dried sewage sludge	56.3 ±0.20	4.44 ±0.04	7.24 ±0.05	0.26 ±0.02	2.26 ±0.06	0.39 ±0.05	90.7 ±0.6	66.1 ±5.3	29.8 ±0.1	1.88 ±0.03	4.53 ±1.02
Poultry litter ash	-	0.03 ±0.01	18.1 ±0.8	24.0 ±0.8	9.68 ±0.47	6.27 ±0.03	110 ±8	122 ±3	19.1 ±0.4	5.27 ±0.10	0.24 ±0.01

OM – organic matter content

Sewage sludge based fertilizer production

In the investigated technology sewage sludge, poultry litter ash and conventional fertilizers were mixed and granulated with diluted mineral acids HNO_3 or H_2SO_4. Sewage sludge was used as a source of essential organic compounds and slow release nutrients [88]. Poultry litter ash was added to enrich fertilizers in phosphorus and potassium. The direct application of poultry litter ash as well as the use as a substitute in fertilizer production by mixing with potassium chloride and triple superphosphate are a fairly common practice in Europe [74]. Such mineral fertilizers as potassium sulphate, potassium nitrate, ammonia nitrate, ammonia sulphate and diammonium phosphate was added in order to increase primary nutrient content of final products and to obtain correct NPK ratio. Mineral acids play role of bending and disinfecting agent and increase the bioavailability of nutrients.

Based on the composition of sewage sludge and poultry litter ash a number of organo-mineral fertilizers were made containing 46-63 % of secondary raw materials. The amount of input materials was selected in such a way as to obtain the possible maximum content of waste in final product and at the same time do not excess permissible concentration of heavy metals. The second purpose was to modify the composition of products by mineral fertilizers in order to enrich and correct NPK ratio in respect of plant requirements (rape, flax, sunflower). The composition of sewage sludge based fertilizers is covered by know-how.

6. Results and discussion

The amount of primary nutrient in sewage sludge based fertilizers is presented in *Table 6. NPK content of sewage sludge based fertilizers*Table 6. The average sum of NPK expressed as $N+P_2O_5+K_2O$ in final products is 30.3%, 33.3% and 31.8% in fertilizers for rape, flax and sunflower crops respectively. NPK ratio in organo-mineral fertilizers for flax and sunflower crops is in the rage of nutritional requirements according to [89]. In case of fertilizers for rape crop the average nitrogen content is slightly lower than required, which can be attributed to the nitrogen losses during the granulation process.

Table 6. NPK content of sewage sludge based fertilizers

		Fertilizers for rape crop	**Fertilizers for flax crop**	**Fertilizers for sunflower crop**
N	% DM	Range 7.93-11.9 Mean 10.6 Median 11.2 SD 1.84	Range 4.33-7.11 Mean 5.46 Median 5.20 SD 1.18	Range 5.71-7.59 Mean 6.59 Median 6.53 SD 0.88
P_2O_5	% DM	Range 4.32-4.81 Mean 4.65 Median 4.73 SD 0.22	Range 8.87-13.3 Mean 10.7 Median 10.3 SD 2.07	Range 6.48-9.64 Mean 8.49 Median 8.91 SD 1.38
K_2O	% DM	Range 12.3-16.5 Mean 15.0 Median 15.6 SD 1.93	Range 15.3-19.4 Mean 17.1 Median 16.9 SD 2.0	Range 16.0-17.8 Mean 16.7 Median 16.6 SD 0.7
Average NPK* ratio in fertilizers		2.4 : 1 : 3.3	1 : 3 : 3.3	1 : 1.4 : 2.5
Plant required NPK* ratio		2.8-3.3 : 1-1.4 : 3.3-4.4	1-1.3 : 2-2.7 : 3-4	1-1.3 : 1-1.5 : 2-3

NPK – mass ratio $N:P_2O_5:K_2O$

Obtained fertilizers generally are characterized by low content of water soluble phosphorus. It can be attributed to the fact, that the most of phosphorus (54-100%) are derived from secondary raw materials, which contain P in slightly water soluble forms. In sewage sludge phosphorus is presented in organic form and inorganic, usually as Fe-, Al- and Ca-phosphates. The proportion of this forms is dependent on treatment processes used in WWTPs [90]. In case of poultry litter ash phosphorus has been found as calcium phosphates and apatite [61]. However, field experiments showed, that phosphorus availability in sewage sludge and poultry litter ash can significantly increase after application and is determined by soil forming factors and processes occurring [91, 92]. The concentration of water soluble phosphorus in fertilizers for flax and sunflower crops are higher in comparison with fertilizers for rape crop. This results from DAP content in composition of fertilizers for flax and sunflower crops. From the literature data follows, that only 10-15% of phosphorus is taking up by plant during first year of fertilization, the rest can be fixed in forms, which are not available for plant or lost to groundwater [93]. From this point of view, low content of water soluble phosphorus in sewage based fertilizers seems to be an advantage in comparison with conventional fertilizers. The content of phosphorus soluble in 2% citric acid in sewage sludge based fertilizers is relatively high and varies from 63.6% of total phosphorus content in case of fertilizers for sunflower crop to 96.7% in case of fertilizers dedicated for flax crop. It should be noted, that sewage sludge based fertilizers have higher content of potentially available phosphorus (by 36-91%) in comparison with sewage sludge, which is associated not only with DAP content in fertilizers dedicated to flax and sunflower crops, but also with acid addition during granulation process.

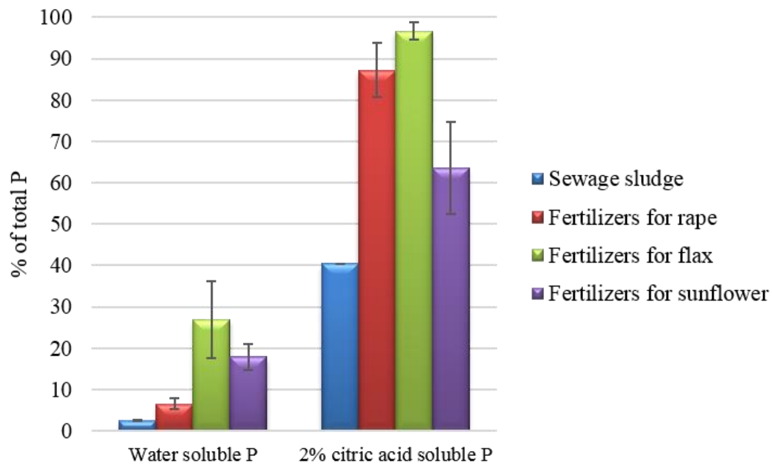

Figure 4. Phosphorus bioavailability of sewage sludge and sewage sludge-based fertilizers (the error bars in case of fertilizers mean the amplitude of the average values)

All obtained organo-mineral fertilizers fulfill the requirements regarding heavy metal content according to polish legislation (Table 7). It should be noted, that Ni and Cr content in final products is relatively high, which results from high concentration of this metals in sewage sludge and poultry litter ash. Reduction of heavy metals content in fertilizers can be achieved through substitution of some amount of secondary raw materials by mineral fertilizers. However, the conventional fertilizers also can contain high Ni and Cr content [94]. The point is that concentration of this metals in mineral fertilizers are not regulated by directive. Therefore, fertilization by mineral fertilizers contributes to accumulation of significant amount of Ni and Cr in soil.

Table 7. Heavy metal content of sewage sludge based fertilizers

		Fertilizers for rape crop	**Fertilizers for flax crop**	**Fertilizers for sunflower crop**	**Polish fertilizer standards for organo-mineral fertilizers** [95]	**Polish mineral NPK fertilizers (n*=19)** [94]
Cr	mg/kg DM	Range 46.8-62.1 Mean 50.9 Median 47.4 SD 7.50	Range 65.2-97.8 Mean 76.1 Median 70.7 SD 15.3	Range 59.5-75.4 Mean 66.2 Median 64.9 SD 6.7	100	Range 38.7-168.7 Mean 87.2 Median 72.5 SD 32.2
Pb		Range 14.8-18.9 Mean 17.3 Median 17.8 SD 1.76	Range 16.7-23.8 Mean 20.2 Median 20.1 SD 3.9	Range 17.2-21.8 Mean 20.0 Median 20.5 SD 2.2	140	Range 0.50-5.0 Mean 2.31 Median 2.17 SD 1.30
Ni		Range 53.1-58.4 Mean 56.0 Median 56.2 SD 2.18	Range 52.0-58.3 Mean 55.3 Median 55.5 SD 3.09	Range 56.1-59.0 Mean 56.9 Median 57.2 SD 2.3	60	Range 7.60-396 Mean 102 Median 68.5 SD 87.7
Cd		Range 1.35-2.12 Mean 1.68 Median 1.62 SD 0.322	Range 1.72-2.06 Mean 1.88 Median 1.87 SD 0.14	Range 0.53-1.22 Mean 0.86 Median .85 SD 0.35	5	Range 2.90-12.3 Mean 5.62 Median 4.96 SD 2.43
Hg		Range 0.286-0.670 Mean 0.495 Median 0.512 SD 0.192	Range 0.556-0.894 Mean 0.716 Median 0.707 SD 0.161	Range 0.288-1.32 Mean 0.732 Median 0.679 SD 0.544	2	Range 0.017-0.258 Mean 0.056 Median 0.036 SD 0.054

n – number of samples

Average compressive strength of sewage sludge based fertilizers are vary from 2.79±1.47 N/granule in case of fertilizers for flax crop to 127.4±34.7 N/granule in case of fertilizers for rape crop. The proper value of compressive strength of fertilizer granules to withstand normal handling and storage is more than 20 N/granule. Only fertilizers for rape crop fulfil this requirement. According to [96, 97] compressive strength of organo-mineral fertilizer granules depends on water, organic matter and iron content. However, no deference in this parameters in sewage sludge based fertilizers are observed. The optimization of granulation process, especially selection of solid/liquid ratio is required to enable production of fertilizers for flax and sunflower crops with high compressive strength of granules.

To assess the potential value of investigated technology the SWOT analysis was used (Table 8). The technology is an example of zero-waste method. It is not include complicated technological operations and do not require high investment cost. Fertilizer production from secondary raw materials partial overcomes the problem with waste management, and gives a possibility of nutrient and organic matter recycling, which is associated with efficient natural resource use. The obtained fertilizers characterized by high nutrient content, are safe, stable and easy for application. However, the emphasis should be placed on quality of input materials. Only treated sewage sludge can be used for fertilizer production in order to not pose a threat to the environment. Heavy metal content of secondary raw materials also should be taken into account. One of the main drawback of this solution is that such fertilizers do not have entry on external European market. There is due to the fact, that in newer fertilizer regulation from 2022 sewage sludge was not classified as component material category unlike poultry litter ash [98]. Additionally, the consumer confidence in such products is usually low. The demand for fertilizers based on secondary raw materials is depend on a wide range on various factors. One important factor is a profitability of using these materials in comparison with conventional fertilizers. Costs of waste-based fertilizers including transport and spreading costs, legislation, availability of such products, geographic location, reputation, as well as farmer preferences and perceptions (odors, uncertainty of crop response) will have impact on market development for waste-based fertilizers [99].

Table 8. SWOT analysis of technology for sewage sludge derived fertilizer production

STRENGTS	WEAKNESSES
In line with circular economy Zero-waste method Low investment costs Simplicity of technology Based on local and renewable input materials Product with high nutrient content Product characterized by slow nutrient release Stable, easy for storage, transport and application product Product offers soil enrichment in organic matter	Variable composition of sewage sludge and poultry litter ash Monitoring of input materials quality is needed No possibility for entering on external European fertilizer market (with current regulation) High competition on fertilizer market
OPPORTUNITIES	THREATS
Nutrient recycling Organic matter recycling Reducing of mineral fertilizer consumption Partial management of sewage sludge and poultry litter ash Reducing of nutrient leakage	Potential presence of pollutants Lack of consumer confidence Lack of mechanisms and incentives Difficulties related to waste status of input materials (sewage sludge, poultry litter ash)

9. Conclusions

Pressure on natural resources and environmental degradation force to take actions towards a more sustainable economic model. The new European Union policy is aimed at optimizing the use of resources and energy and reduce the amount of waste. Taking into account nutrient content, pollutant presence and possibility of treatment technologies, sewage and manure are considered the promising waste streams for nutrient recycling and recovery. Available on the market technology for nutrient recovery usually are focused only on phosphorus as non-renewable resource. However, it is a need of total value recovery from waste including organic matter and nutrients in line of circular economy.

The technology of fertilizer production from sewage sludge ad poultry litter ash opens the opportunity of transformation waste into valuable products. In order to meet the requirements in respect of heavy metal content in fertilizers the right amount of input materials should be selected. It is possible to modify the composition of final products by adding mineral fertilizers to obtain fertilizers with correct NPK ratio depends on nutritional requirements of various crops.

The organo-mineral fertilizers for rape, flax and sunflower crops were produced. The obtained fertilizers characterized by high nutrient content (>30%) and heavy metal content, which did not excess the permissible concentration settled up by legislation. It is calculated, than 16-39% of N, 54-100% of P_2O_5 and 16-29% of K_2O in organo-mineral fertilizers came from secondary raw materials. Such substitution can significantly reduce costs of fertilizer production and manage a large volume of waste (maximum waste content in fertilizers 63%) in environmental friendly way.

References

[1] United Nations Department of Economic and Social Affairs Population Division, “World Population Prospects: The 2017 Revision, Key Findings and Advance Tables,” ESA/P/WP/248, 2017.

[2] Food and Agriculture Organisation of the United Nations, *More people, more food, worse water ? a global review of water pollution from agriculture*. 2018.
[3] S. Belboom, C. Szöcs, and A. Léonard, "Environmental impacts of phosphoric acid production using di-hemihydrate process: A Belgian case study," *J. Clean. Prod.*, vol. 108, pp. 978–986, 2015.
[4] S. M. Jasiński, "U.S. Geological Survey. Mineral Commodity Summaries. Phosphate rock." 2018.
[5] M. C. Mew, G. Steiner, and B. Geissler, "Phosphorus supply chain-scientific, technical, and economic foundations: A transdisciplinary orientation," *Sustain.*, vol. 10, no. 4, 2018.
[6] M. Smol, "The importance of sustainable phosphorus management in the circular economy (CE) model: the Polish case study," *J. Mater. Cycles Waste Manag.*, pp. 1–9, 2018.
[7] European Commission, "20 critical raw materials - major challenge for EU industry," no. May, 2014.
[8] Stephen M. Jasiński, "U.S. Geological Survey. Mineral Commodity Summaries. Potash." 2018.
[9] D. J. Batstone, T. Hülsen, C. M. Mehta, and J. Keller, "Platforms for energy and nutrient recovery from domestic wastewater: A review," *Chemosphere*, vol. 140, pp. 2–11, 2015.
[10] M. K. Winkler and L. Straka, "New directions in biological nitrogen removal and recovery from wastewater," *Curr. Opin. Biotechnol.*, vol. 57, pp. 50–55, 2019.
[11] F. Brentrup and C. Palliere, "Energy efficiency and greenhouse gas emissions in European nitrogen fertilizer production and use," 2008.
[12] A. Buckwell and E. Nadeu, *Nutrient Recovery and Reuse (NRR) in European agriculture. A review of the issues, opportunities, and actions*. Brussels: RISE Foundation, 2016.
[13] European Commission, "Resource Efficiency in Practice – Closing Mineral Cycles," 2016.
[14] B. Su, A. Heshmati, Y. Geng, and X. Yu, "A review of the circular economy in China: Moving from rhetoric to implementation," *J. Clean. Prod.*, vol. 42, pp. 215–227, 2013.
[15] A. Heshmati, "a Review of the Circular Economy and Its Implementation En Treprenörsk a Psforum," no. 9611, 2015.
[16] European Commission, *Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. Towards a circular economy: A zero waste programme for Europe*. 2014.
[17] J. Kirchherr, D. Reike, and M. Hekkert, "Conceptualizing the circular economy: An analysis of 114 definitions," *Resour. Conserv. Recycl.*, vol. 127, pp. 221–232, 2017.
[18] B. K. Mayer *et al.*, "Total Value of Phosphorus Recovery," *Environ. Sci. Technol.*, vol. 50, no. 13, pp. 6606–6620, 2016.
[19] P. Dorota, "Environmental aspects of managing the organic matter in agriculture," *Econ. Reg. Stud.*, vol. 8, no. 2, pp. 98–112, 2015.
[20] Eurostat, "Sewage sludge production in European Union." [Online]. Available: http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=env_ww_spd&lang=en. [Accessed: 13-Feb-2019].
[21] J. Colón *et al.*, "Producing sludge for agricultural applications," in *Innovative wastewater treatment and resource recovery technologies*, 2017, pp. 292–314.
[22] Q. Liu *et al.*, "Phosphorus speciation and bioavailability of sewage sludge derived biochar amended with CaO," *Waste Manag.*, vol. 87, pp. 71–77, 2019.
[23] A. Zaker, Z. Chen, X. Wang, and Q. Zhang, "Microwave-assisted pyrolysis of sewage sludge: A review," *Fuel Process. Technol.*, vol. 187, pp. 84–104, 2019.
[24] K. Gondek, M. Kopeć, and G. Tomasz, "Sewage sludge as a source of microelements regarding fodder value of spring wheat grain," *Ecol. Chem. Eng.*, vol. 20, no. 9, pp. 975–985, 2013.
[25] Y. Bai *et al.*, "Sewage sludge as an initial fertility driver for rapid improvement of mudflat salt-soils," *Sci. Total Environ.*, vol. 578, pp. 47–55, 2017.
[26] S. Mattana *et al.*, "Sewage sludge processing determines its impact on soil microbial community structure and function," *Appl. Soil Ecol.*, vol. 75, pp. 150–161, 2014.
[27] S. D. Koutroubas, V. Antoniadis, S. Fotiadis, and C. A. Damalas, "Growth, grain yield and nitrogen use efficiency of Mediterranean wheat in soils amended with municipal sewage sludge," *Nutr. Cycl. Agroecosystems*, vol. 100, pp. 227–243, 2014.
[28] E. Ociepa, M. Mrowiec, and J. Lach, "Influence of fertilisation with sewage sludge-derived preparation on selected soil properties and prairie cordgrass yield," *Environ. Res.*, vol. 156, no. July 2016, pp. 775–780, 2017.
[29] N. Roig, J. Sierra, E. Martí, M. Nadal, M. Schuhmacher, and J. L. Domingo, "Long-term amendment of Spanish soils with sewage sludge: Effects on soil functioning," *Agric. Ecosyst. Environ.*, vol. 158, pp. 41–48, 2012.
[30] Eurostat, "Methods of sewage sludge disposal in European Union." [Online]. Available: https://ec.europa.eu/eurostat/tgm/refreshTableAction.do?tab=table&plugin=1&pcode=ten00030&language=en. [Accessed: 13-Feb-2019].
[31] M. Praspaliauskas and N. Pedišius, "A review of sludge characteristics in Lithuania's wastewater treatment plants and perspectives of its usage in thermal processes," *Renew. Sustain. Energy Rev.*, vol. 67, pp. 899–907, 2017.
[32] C. B. Rizzardini and D. Goi, "Sustainability of domestic sewage sludge disposal," *Sustain.*, vol. 6, pp. 2424–2434, 2014.
[33] V. Jones, M. Gardner, and B. Ellor, "Concentrations of trace substances in sewage sludge from 28 wastewater treatment works in the UK," *Chemosphere*, vol. 111, pp. 478–484, 2014.
[34] P. Alvarenga *et al.*, "Sewage sludge, compost and other representative organic wastes as agricultural soil amendments: Benefits versus limiting factors," *Waste Manag.*, vol. 40, pp. 44–52, 2015.
[35] M. Tytła, K. Widziewicz, and E. Zielewicz, "Heavy metals and its chemical speciation in sewage sludge at different stages of processing," *Environ. Technol. (United Kingdom)*, vol. 37, no. 7, pp. 899–908, 2016.
[36] F. Bastida, N. Jehmlich, J. Martínez-Navarro, V. Bayona, C. García, and J. L. Moreno, "The effects of struvite and sewage sludge on plant yield and the microbial community of a semiarid Mediterranean soil," *Geoderma*, vol. 337, pp. 1051–1057, 2019.
[37] T. Spanos, A. Ene, C. Styliani Patronidou, and C. Xatzixristou, "Temporal variability of sewage sludge heavy metal content from Greek wastewater treatment plants," *Ecol. Chem. Eng. S*, vol. 23, no. 2, pp. 271–283, 2016.
[38] B. O. Clarke and S. R. Smith, "Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids," *Environ. Int.*, vol. 37, no. 1, pp. 226–247, 2011.
[39] E. Z. Harrison, S. R. Oakes, M. Hysell, and A. Hay, "Organic chemicals in sewage sludges," *Sci. Total Environ.*, vol. 367, pp. 481–497, 2006.
[40] A. A. Al-Gheethi, A. N. Efaq, J. D. Bala, I. Norli, M. O. Abdel-Monem, and M. O. Ab. Kadir, "Removal of pathogenic bacteria from sewage-treated effluent and biosolids for agricultural purposes," *Appl. Water Sci.*, vol. 8, no. 74, p. 25, 2018.
[41] K. Bibby and J. Peccia, "Identification of viral pathogen diversity in sewage sludge by metagenome analysis," *Environ. Sci. Technol.*, vol. 47, no. 4, pp. 1945–1951, 2013.
[42] European Commission, *Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC)*. 1986.

[43] G. Mininni, A. R. Blanch, F. Lucena, and S. Berselli, "EU policy on sewage sludge utilization and perspectives on new approaches of sludge management," *Environ. Sci. Pollut. Res.*, vol. 22, no. 10, pp. 7361–7374, 2015.

[44] M. Zennegg, M. Munoz, P. Schmid, and A. C. Gerecke, "Temporal trends of persistent organic pollutants in digested sewage sludge (1993-2012)," *Environ. Int.*, vol. 60, pp. 202–208, 2013.

[45] H. Kirchmann, G. Börjesson, T. Kätterer, and Y. Cohen, "From agricultural use of sewage sludge to nutrient extraction: A soil science outlook," *Ambio*, vol. 46, pp. 143–154, 2017.

[46] B. Wiechmann, C. Dienemann, C. Kabbe, S. Brandt, I. Vogel, and A. Roskosch, "Sewage Sludge Management," 2013.

[47] H. Bauman-Kaszubska and M. Sikorski, "Charakterystyka ilościowa i jakościowa osadów ściekowych pochodzących z małych oczyszczalni ścieków w powiecie Płockim," *Inżynieria Ekol.*, no. 25, pp. 20–29, 2011.

[48] M. C. Samolada and A. A. Zabaniotou, "Comparitive assesment of municipal sewage sludge incineration, gasification and pyrolysis for a sustainable sludge-to-energy management in Greece," *Waste Manag.*, vol. 34, pp. 411–420, 2014.

[49] M. Atienza-Martínez, G. Gea, J. Arauzo, S. R. A. Kersten, and A. M. J. Kootstra, "Phosphorus recovery from sewage sludge char ash," *Biomass and Bioenergy*, vol. 65, pp. 42–50, 2014.

[50] L. Świerczek, B. M. Cieślik, and P. Konieczka, "The potential of raw sewage sludge in construction industry – A review," *J. Clean. Prod.*, vol. 200, pp. 342–356, 2018.

[51] C. J. Lynn, R. K. Dhir, and G. S. Ghataora, "Environmental impacts of sewage sludge ash in construction: Leaching assessment," *Resour. Conserv. Recycl.*, vol. 136, no. February, pp. 306–314, 2018.

[52] S. S. A. Syed-Hassan, Y. Wang, S. Hu, S. Su, and J. Xiang, "Thermochemical processing of sewage sludge to energy and fuel: Fundamentals, challenges and considerations," *Renew. Sustain. Energy Rev.*, vol. 80, pp. 888–913, 2017.

[53] L. Fang *et al.*, "Phosphorus recovery and leaching of trace elements from incinerated sewage sludge ash (ISSA)," *Chemosphere*, vol. 193, pp. 278–287, 2018.

[54] K. Gorazda *et al.*, "Fertilisers production from ashes after sewage sludge combustion – A strategy towards sustainable development," *Environ. Res.*, vol. 154, pp. 171–180, 2017.

[55] A. Kelessidis and A. S. Stasinakis, "Comparative Study of the Methods Used for the Treatment and Final Disposal of Sewage Sludge in European Countries," no. September, pp. 8–10, 2011.

[56] M. P. Bernal *et al.*, "Evaluation of manure management systems in Europe," 2015.

[57] M. P. Bernal *et al.*, "Evaluation of manure management systems in Europe," 2015.

[58] D. Huygens, H. Saveyn, D. Tonini, P. Eder, and L. D. Sancho, "Pre-final STRUBIAS Report. DRAFT STRUBIAS recovery rules and market study for precipitated phosphate salts&derivates, thermal oxidation materials&derivates and pyrolysis&gasification materials in view of their possible inclusion as Component Material Cate."

[59] H. L. Foged, X. Flotats, A. Bonmati, J. Palatsi, A. Magri, and K. M. Schelde, "Inventory of manure processing activities in Europe. Technical Report No. I concerning 'Manure Processing Activities in Europe' to the European Commission," 2011.

[60] S. Marttinen *et al.*, "Towards a breakthrough in nutrient recycling. State-of-the-art and recommendations for developing policy instruments in Finland," 2018.

[61] E. Brod, "Manure-based recycling fertilisers. A literature review of treatment technologies and their effect on phosphorus fertilization effects," 2018.

[62] E. Bloem *et al.*, "Contamination of organic nutrient sources with potentially toxic elements, antibiotics and pathogen microorganisms in relation to P fertilizer potential and treatment options for the production of sustainable fertilizers: A review," *Science of the Total Environment*, vol. 607–608. Elsevier B.V., pp. 225–242, 2017.

[63] *Regulation (EC) No 1069/2009 of the European Parliament and of the Council of 21 October 2009 laying down health rules as regards animal by-products and derived products not intended for human consumption and repealing Regulation (EC) No 1774/2002 (Animal*, vol. 2009. 2009, pp. 1–33.

[64] M. Sarvi, K. Ylivainio, and E. Turtola, "Report on environmentally relevant heavy metals in P-fertilizer materials," 2017.

[65] F. Zhang, Y. Li, M. Yang, and W. Li, "Content of heavy metals in animal feeds and manures from farms of different scales in Northeast China," *Int. J. Environ. Res. Public Health*, vol. 9, no. 8, pp. 2658–2668, 2012.

[66] T. P. Van Boeckel *et al.*, "Global trends in antimicrobial use in food animals.," *Proc. Natl. Acad. Sci. U. S. A.*, vol. 112, no. 18, pp. 5649–54, 2015.

[67] M. Pan and L. M. Chu, "Transfer of antibiotics from wastewater or animal manure to soil and edible crops," *Environ. Pollut.*, vol. 231, pp. 829–836, 2017.

[68] M. Conde-Cid *et al.*, "Occurrence of tetracyclines and sulfonamides in manures, agricultural soils and crops from different areas in Galicia (NW Spain)," *J. Clean. Prod.*, vol. 197, pp. 491–500, 2018.

[69] European Medicines Agency, "Sales of veterinary antimicrobial agents in 30 European countries in 2015. Trends from 2010 to 2015 Seventh ESVAC report," 2014.

[70] B. Albero, J. L. Tadeo, M. Escario, E. Miguel, and R. A. Pérez, "Persistence and availability of veterinary antibiotics in soil and soil-manure systems," *Sci. Total Environ.*, vol. 643, pp. 1562–1570, 2018.

[71] P. D. Millner, "Manure management," in *Encyclopedia of Meat Sciences*, Second Edi., Elsevier, 2014, pp. 152–156.

[72] Y. Hou *et al.*, "Stakeholder perceptions of manure treatment technologies in Denmark, Italy, the Netherlands and Spain," *J. Clean. Prod.*, vol. 172, pp. 1620–1630, 2018.

[73] AMEC Environment & Infrastracture UK Limited in partnership with BIO Intelligence Service, "Collection and Analysis of Data for the Control of Emissions from the Spreading of Manure," 2014.

[74] D. Huygens, H. Saveyn, D. Tonini, P. Eder, and L. D. Sancho, "Pre-final STRUBIAS Report. DRAFT STRUBIAS recovery rules and market study for precipitated phosphate salts&derivates, thermal oxidation materials&derivates and pyrolysis&gasification materials in view of their possible inclusion as Component Material Cate," 2018.

[75] "Fertilizer consumption in European Union." [Online]. Available: https://www.worldfertilizer.com/special-reports/09022018/europe-in-focus/. [Accessed: 08-May-2019].

[76] J. Webb *et al.*, "Emissions of Ammonia, Nitrous Oxide and Methane During the Management of Solid Manures," in *Agroecology and Strategies for Climate Change*, E. Lichtfouse, Ed. Dordrecht: Springer Netherlands, 2012, pp. 67–107.

[77] L. Egle, H. Rechberger, and M. Zessner, "Overview and description of technologies for recovering phosphorus from municipal wastewater," *Resour. Conserv. Recycl.*, vol. 105, pp. 325–346, 2015.

[78] E. Desmidt *et al.*, "Technology Global Phosphorus Scarcity and Full-Scale P- Recovery Techniques : A Review," *Environ. Sci. Technol.*, vol. 45, no. 4, pp. 336–384, 2015.

[79] H. Zhang *et al.*, "Recovery of phosphorus from dairy manure: A pilot-scale study," *Environ. Technol.*, vol. 36, no. 11, pp. 1398–1404, 2015.

[80] M. S. Romero-Güiza, S. Astals, J. M. Chimenos, M. Martínez, and J. Mata-Alvarez, "Improving anaerobic digestion of pig manure by adding in the same reactor a stabilizing agent formulated with low-grade magnesium oxide," *Biomass and Bioenergy*, vol. 67, pp. 243–251, 2014.

[81] A. Escudero, F. Blanco, A. Lacalle, and M. Pinto, "Struvite precipitation for ammonium removal from anaerobically treated effluents," *J. Environ. Chem. Eng.*, vol. 3, pp. 413–419, 2015.
[82] S. Sengupta, T. Nawaz, and J. Beaudry, "Nitrogen and Phosphorus Recovery from Wastewater," *Curr. Pollut. Reports*, vol. 1, no. 3, pp. 155–166, 2015.
[83] L. Liu, C. Pang, S. Wu, and R. Dong, "Optimization and evaluation of an air-recirculated stripping for ammonia removal from the anaerobic digestate of pig manure," *Process Saf. Environ. Prot.*, vol. 94, pp. 350–357, 2015.
[84] H. Huang, J. Li, B. Li, D. Zhang, N. Zhao, and S. Tang, "Comparison of different K-struvite crystallization processes for simultaneous potassium and phosphate recovery from source-separated urine," *Sci. Total Environ.*, vol. 651, pp. 787–795, 2019.
[85] E. Tarragó, M. Ruscalleda, J. Colprim, M. D. Balaguer, and S. Puig, "Towards a methodology for recovering K-struvite from manure," *J. Chem. Technol. Biotechnol.*, vol. 93, pp. 1558–1562, 2018.
[86] B. M. Cieślik, J. Namieśnik, and P. Konieczka, "Review of sewage sludge management: Standards, regulations and analytical methods," *J. Clean. Prod.*, vol. 90, pp. 1–15, 2015.
[87] H. Kominko, K. Gorazda, and Z. Wzorek, "The Possibility of Organo-Mineral Fertilizer Production from Sewage Sludge," *Waste and Biomass Valorization*, vol. 8, no. 5, pp. 1781–1791, 2017.
[88] Q. Lu, Z. L. He, and P. J. Stoffella, "Land application of biosolids in the USA: A review," *Appl. Environ. Soil Sci.*, vol. 2012, 2012.
[89] *International Fertilizer Industry Association. World fertilizer use manual*. Paris, 1992.
[90] D. Houben, E. Michel, C. Nobile, H. Lambers, E. Kandeler, and M. P. Faucon, "Response of phosphorus dynamics to sewage sludge application in an agroecosystem in northern France," *Appl. Soil Ecol.*, vol. 137, pp. 178–186, 2019.
[91] N. Glæsner, F. van der Bom, S. Bruun, T. McLaren, F. H. Larsen, and J. Magid, "Phosphorus characterization and plant availability in soil profiles after long-term urban waste application," *Geoderma*, vol. 338, pp. 136–144, 2019.
[92] Faridullah, M. Irshad, A. E. Eneji, and Q. Mahmood, "Plant nutrient release from poultry litter and poultry litter ash amended soils by various extraction methods," *J. Plant Nutr.*, vol. 36, no. 3, pp. 357–371, 2013.
[93] T. L. Roberts and A. E. Johnston, "Phosphorus use efficiency and management in agriculture," *Resour. Conserv. Recycl.*, vol. 105, pp. 275–281, 2015.
[94] F. Gambuś J. and Wieczorek, "Pollution of Fertilizers with Heavy Metals," *Ecol. Chem. Eng.*, vol. 19, no. 4–5, pp. 353–360, 2012.
[95] *Rozporządzienie Ministra Rolnictwa i Rozwoju Wsi z dnia 18 czerwca 2008 r. w sprawie wykonania niektórych przepisów ustawy o nawozach i nawożeniu*. Warszawa, 2008.
[96] M. Paré, S. Allaire, L. Khiari, and C. Nduwamungu, "Physical properties of organo-mineral fertilizers--Short Communication.," *Can. Biosyst. Eng.*, vol. 51, pp. 21–27, 2009.
[97] S. E. Allaire and L. E. Parent, "Physical Properties of Granular Organic-based Fertilisers, Part 1: Static Properties," *Biosyst. Eng.*, vol. 87, no. 1, pp. 79–87, 2004.
[98] *Proposal for a regulation of the European Parliament and of the Council laying down rules on the making available on the market of EU fertilizing products and amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009*. 2018, pp. 1–279.
[99] S. Lupton, "Markets for waste and waste–derived fertilizers. An empirical survey," *J. Rural Stud.*, vol. 55, pp. 83–99, 2017.