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Abstract: For satisfying human needs every day, a gradual and inexorable depletion of the Earth occurs. The lack of resources and issues connected with treatment and recovery/disposal of sludge, derived from wastewater treatment plants (WWTPs), are gaining importance internationally. This led to promote the reuse, recycling and recovery of wastes, which are no longer a problem but an opportunity. Furthermore, Directive 2008/98/EC introduces the waste hierarchy that forced the technicians to re-think completely the waste management strategy by preventing landfill disposal. In this context, the present paper sought to review the possible alternative for reuse, recycling and recovery of biological sewage sludge produced in the WWTPs as substitute of natural material. Authors explored the application of biosolids on land, such as amendant/fertilizer both in agriculture and for recovery of degraded sites, and on engineering fields, in partial or total substitution of virgin materials. The recovery of biosolids as adsorbent materials and as a source of phosphorus is also treated.

*Title Page

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Abstract:

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Pavia, October 11th, 2018

Dear Editor-in-Chief,
we would like to submit the manuscript titled "BIOSOLIDS: WHAT ARE THE DIFFERENT TYPES OF RECOVERY?" for possible publication in *Journal of Cleaner Production*.

The present paper sought to review the possible alternative for reuse, recycling and recovery of biological sewage sludge produced in the WWTPs as substitute of natural material. Authors explored the application of biosolids on land, such as amendant/fertilizer both in agriculture and for recovery of degraded sites, and on engineering fields, in partial or total substitution of virgin materials. The recovery of biosolids as adsorbent materials and as a source of phosphorus is also treated.

This review shows that generally population perceives the use of biosolids as dangerous for human and environment while in land application of biosolids, heavy metals and pathogens effects are under study.
Feasibility of the utilization of biosolids for brick construction, conversion into adsorbent material and source of phosphorus is also presented. Further investigation on "emerging" organic contaminants would be conducted.

This paper is original and unpublished; it is not being considered for publication by any other journal.

Yours Sincerely,

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HIGHLIGHTS

- Population perceives the use of biosolids as dangerous for human and environment
- In agricultural reuse of biosolids, heavy metals effects continue to be studied
- Feasibility of the utilization of biosolids for brick construction
- Conversion into adsorbent material and source of phosphorus is also presented
- Further investigation on “emerging” organic contaminants would be conducted

Biosolids: what are the different types of recovery?

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Abstract

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Keywords

water treatment sludge; biosolids recovery; construction products; agricultural land; adsorbent materials

17800 words including tables, figure captions and references.

About 11,000 words with tables and figure captions but without references.

1. Introduction

We live in a system, the Earth, which has finished size; therefore, its space and its resources are limited. Nevertheless, in the last 60 years, population is more than doubled; it increased from 3.5 billion in the 1960s' to 7.3 billion in the 2015s' and it is expected to reach 8.5 billion by 2030 (UN DESA, 2015). Therefore, the energy demand, the intensive use of land increase and the flow of materials to meet our consumption increase; so, the production of wastes grows.

For satisfying their needs, humans mainly used non-renewable resources (at the moment, worldwide energy demand is mainly satisfied by means of fossil fuels (Siddiquee and Rohani, 2011); therefore, every day, a gradual and inexorable depletion of the Earth occurs. Depletion is not referred only to the consumption of natural resources but also to the ecosystem; for instance, the mining of geologic materials alters habitats, causes increased runoff and soil erosion, and disrupts the ecological processes of the land where the mining occurs. Moreover, reduced forest cover from mining may negatively affect the planet's ability to process CO_2 (Calkins, 2009).

In view of the considerations reported above, it is obvious that we need to rethink the way for using resources, moving towards a system where resources are managed in a sustainable way *id est* "a development that meets the needs of the present without compromising the ability of future generations to meet their own needs" (Butlin, 1989).

The simultaneous requirement to manage resources and wastes in a more rational way has meant that many communities worldwide have begun to search for long-term alternative solutions instead methods employed to dispose of their waste and to produce energy. Therefore, people are involved in an epochal paradigm shift: wastes are no longer a problem but an opportunity.

In the EU, for instance, the first step was done about 20 years ago, when it was approved the so-called Landfill Directive (CEU, 1999) although the change of direction was then established with the Waste Framework Directive – WFD (EP/CEU, 2008). The WFD sets the basic concepts and definitions related to waste management (i.e. definitions of waste, recycling, recovery) and explains when waste ceases to be waste and becomes a secondary raw material (it is known as end-of-waste criteria). Moreover, it introduces the waste hierarchy that forced the technicians to re-think completely the waste management strategy because landfill disposal can be done only for wastes that cannot be reused, recycled and recovered.

In order to promote the reuse, recycling and recovery of wastes, the European Commission adopted an ambitious *Circular Economy Package*. Aim of circular economy is closing the loop of product lifecycles keeping their added value for as long as possible and eliminate waste with obvious benefits for the environment and the economy. In order to demonstrate that the transition towards a more circular economy is feasible, a research project is financed within the

EU Research and Innovation programme - Horizon 2020. In this context, the management of the residues produced by the wastewater treatment process is further complicated by the Urban Waste Water Treatment Directive 91/271/EC (UWWTD) (CEU, 1991) because, at the same time, the wastewater treatment plants (WWTPs) have to: (1) collect and treat more polluted water; (2) satisfy the more stringent effluent quality standards foreseen by the UWWTD; (3) satisfy the waste hierarchy introduced by the WFD. However, as known, a better water treatment efficiency involves greater production of sludge with higher level of contamination (Mininni et al., 2015). Therefore, in the last years, researches are mainly focused on the development of technologies/management strategies aimed at preventing the sludge production. Details on sludge minimization techniques can be found, for instance, in: (Collivignarelli et al., 2014; M.C. Collivignarelli et al., 2017, 2015; Maria Cristina Collivignarelli et al., 2017; Foladori et al., 2010; Paul and Liu, 2012).

2. Methods and Scope

Aim of this work is exploring the potential reuse and recovery of sewage sludge as substitute of natural materials. A comprehensive review of different methods reported in peer-reviewed journals, conference proceedings, published reports and other documents presenting, sustainable sludge management through recovery, recycling and reuse, has been done to prepare this document.

The review consists of five sections; the first one (general overview) reports the main chemical-physical characteristics of biosolids obtained by literature from all over the world. In the second one, instead, the following application are explored: biosolids on land, such as amendment/fertilizer both in agriculture and for recovery of degraded sites focusing mainly on reports from USA, New Zealand and Europe since scientists from those regions show high interest in the presented subject. In the third section the recovery of biological sewage sludge in the engineering fields are exposed, *id est* where sludge is utilized in partial or total substitution of raw materials (e.g. bricks and cement production, road construction, etc.), with studies published in technical journals and books. In the fourth section, the applications of biosolids as adsorbent material and as a source of phosphorus are reported. Finally, a brief discussion on the main advantages and drawbacks on biological sewage sludge recovery/reuse.

This review does not only highlight the environmental aspects related to the possible reuse of biosolids, but also focuses on the technical-engineering and health aspects deriving from it.

3. General overview on sludge characteristics

Sludge resulting from wastewater treatment operations and processes is usually in the form of a liquid or semisolid liquid that typically contains from 0.25 to 20% solids by weight, depending on the operations and processes used

(Tchobanoglous et al., 2003). Generally, the term "sludge" refers to a liquid (contains up to 8-10 wt% of dry solids) that does not submitted to further treatments. On the contrary, with the term "biosolids" it is indicated a sludge that had received one or more treatments, which can be: aerobic or anaerobic digestion, alkaline stabilization, thermal drying, acid oxidation/disinfection, composting, etc. (Ukwatta et al., 2015). Therefore, sludge characteristics are mainly affected by the chemical-physical characteristics of influent, which depends on the sewer characteristics (combined or separated sewer) and the presence of industrial activities in the urban aggregate as well as WWTP configuration (e.g. presence of primary settler, anaerobic stabilization stage, etc.).

According with WWTP configuration, different kinds of sludge can be found: primary, secondary (also called biological or waste activated sludge – WAS), mixed, tertiary (which is produced when advanced wastewater treatments are used for removing suspended and dissolved substances remained after conventional biological (secondary) treatment) and digested sludge.

Primary sludge is the residue deriving from primary settlers; it is mainly composed by readily settleable solids and floating material contained in the raw wastewater. Generally, primary sludge is characterised by high putrescibility and a content of total solids (TS) ranging from 2 to 7 wt% (Madlool et al., 2011; Tchobanoglous et al., 2003; Turovskiy and Mathai, 2006). *Secondary sludge* is the residue produced during the biological treatment (i.e. activated sludge process or biofilm systems) of wastewater. It is a complex heterogeneous mixture of microorganisms, bacterial constituents (nucleic acids, proteins, carbohydrates and lipids) (Manara and Zabaniotou, 2012) undigested organics (e.g. paper, plant residues, oils, faecal material, etc.), inorganic materials (not removed in the primary basin, if any), and water (Bianchini et al., 2015; M. C. Samolada and Zabaniotou, 2014). These substances, which are initially in the liquid phase of wastewater, can be found within the sludge in suspended or dissolved form. The wastewater treatment trains will determine the different quantities of the sludge inorganic compounds, and thus the extent to which those compounds are associated to the sludge organic fraction (Tchobanoglous et al., 2003). Generally, TS content ranges from 0.5 to 1.5 wt% (Tchobanoglous et al., 2003; Turovskiy and Mathai, 2006). More details on secondary sludge characteristics are reported in Table 1.

Mixed sludge is obtained when different kinds of sludge (e.g. primary, secondary and tertiary) are mixed; its characteristics depend on the sludge characteristics mixed.

According with the size of WWTPs, sludge produced in the wastewater treatment trains can be submitted to additional treatments in the so-called sludge treatment trains. Aims of those treatments are to reduce the water content (e.g. thickening) and/or stabilize the organic matter (mainly anaerobic digestion, aerobic stabilization). After those stages, thickened/stabilized sludge is submitted to a dewatering process (centrifugation, belt and filter presses are the widely used) in order to obtain a solid residue (in that case called biosolid) with a TS content ranging from 12 to 30% by weight (Bianchini et al., 2015; Sanin et al., 2011).

In Table 1 the main properties of municipal and industrial sludge are summarized; detailed considerations about organic contaminants are reported in the section concerning the recovery of biosolids in agriculture.

Table 1: *Main characteristics of municipal and industrial sludge useful for evaluating recovery and reuse options*

Parameters	**Sludge**		**References**
	Municipal	**Industrial**	
Arsenic (mg kg_{DM}^{-1})	1-10	5-30	(Wang et al., 2008)
Cadmium (mg kg_{DM}^{-1})	1-20	0.5-5	(Wang et al., 2005)
Chromium (mg kg_{DM}^{-1})	1-500	5-2000	(Fuentes et al., 2004)
Copper (mg kg_{DM}^{-1})	100-400	20-2000	(Kim and Owens, 2010)
Lead (mg kg_{DM}^{-1})	20-200	10-50	(Fytili and Zabaniotou, 2008)
Manganese (mg kg_{DM}^{-1})	40-400	5-3000	(Ahmad et al., 2016)
Mercury (mg kg_{DM}^{-1})	1-10	0.2-10	(Tchobanoglous et al., 2003)
Nickel (mg kg_{DM}^{-1})	20-100	15-250	(Maria Cristina Collivignarelli et al., 2015)
Zinc (mg kg_{DM}^{-1})	500-2000	30-1600	(Shamuyarira and Gumbo, 2014)
Chlorine (mg kg_{DM}^{-1})	800-2500	500-3000	(Jordán et al., 2005)
Nitrogen (% of TS)	1.6-6	1-3	(Alvarenga et al., 2015)
Phosphorus (% of TS)	0.5-4	5-20	(Tyagi and Lo, 2011)
Carbon (% of TS)	40-75	35-70	(Praspaliauskas and Pedišius, 2017)
Hydrogen (% of TS)	5-10	5-10	(Murakami et al., 2009)
Oxygen (% of TS)	25	30-40	(Elled et al., 2007)
Phosphorus pentoxide (% of TS)	5-30	5-30	(Kanari et al., 2016)
Sulphur (% of TS)	0.8-1	1-1.5	(Roy et al., 2011)
Moisture (%)	75-99	65-99	(Nomeda et al., 2008)
pH	6.5-7.5	6.4-7.4	(Cheng et al., 2016)

DM: dry matter

As concern heavy metals, from Table 1 it is clear that their content in the sludge is very variable and affected, as stated above, by water and sludge treatment trains and by influent characteristics. Usually sewage sludge deriving from wide urban areas, with a substantial industrial influence, shows higher concentrations of metals, especially Cr and Ni, typically of factories (Jordán et al., 2005). High concentrations of iron (III) and aluminium, instead, can be found in the sewage sludge due to the addition of these salts for favouring, for instance, the phosphorus precipitation (Manara and Zabaniotou, 2012). However, the content of heavy metals in the sludge is a key factor for its reuse and recycling because, as reported in (Cusidó and Cremades, 2012), they can be a threat for human health (for instance, arsenic is carcinogenic; cadmium probably is carcinogenic, teratogenic and embryotoxic; mercury is teratogenic).

Finally, sewage sludge contains some ingredients with agricultural value, such as: organic matter, nitrogen, phosphorus, potassium and, to a lesser extent, calcium, sulphur and magnesium (Jordán et al., 2005; Manara and Zabaniotou, 2012; Tchobanoglous et al., 2003).

Energy content of sewage sludge is usually expressed by means of the Low Heat Value (LHV). LHV value mainly depends on: (i) moisture and ash content (high content involves low LHV); (ii) the undigested organic matter (Fonts et al., 2012; Zhang et al., 2015); (iii) the amount of oxygen (high oxygen content leads to low LHV) (Zhang et al., 2015). As reported in (Stasta et al., 2006) and (Manara and Zabaniotou, 2012), dry sewage sludge has a calorific value similar to that of brown coal (14.6-26.7 MJ kg^{-1}); therefore, sewage sludge can be considered suitable as fossil fuel substitute.

4. Biosolids recovery in agriculture and degraded sites

Land application involves the spreading, spraying, injection, or incorporation of biosolids, including a material derived from biosolids (e.g. compost and pelletized biosolids), onto or below the surface of the land (USEPA, 1995)

Land application of biosolids can represents an interesting strategy for improving site productivity by increasing soil organic matter (SOM) content and fertility; moreover, biosolids can also improve soil physical properties, particularly when applied to heavy textured and poorly structured soils (Alvarenga et al., 2015; Aranda et al., 2015; Castán et al., 2016).

The advantages related to the application of biosolids are well known: mainly, they: (a) improve soil structure, (b) decrease bulk density, (c) increase soil porosity (d) increase soil moisture retention, (e) and hydraulic conductivity (Ojeda et al., 2003) (Figure 1). In addition, thanks to the nitrogen and phosphorus content, biosolids can significantly increase crop yield (Sigua et al., 2005; Wang et al., 2006). In Nelson (New Zealand), for example, aerobically digested biosolids were applied to more than 1000 ha of pine plantation forestland with low soil nitrogen fertility. Results from a long-term trial showed that the application of biosolids has significantly improved forest productivity (Kimberley et al., 2004; Wang et al., 2006) with minimal adverse effects on the ecosystem (Su et al., 2008; Wang et al., 2004). Moreover, as reported in (Wang et al., 2006) applying biosolids to nutrient-deficient plantation forestland can reduce the risk of contaminants entering in the human food chain and it can increase tree growth.

Mine site rehabilitation is another opportunity for sewage sludge recovery. In these cases biosolids are used for restoring contaminated soil (e.g. heavy metals) and mine areas. In Table 2 some recent experiences are summarized.

Table 2: *Examples of biosolids recovered in degraded areas*

Problems	**Sludge applied**	**Results**	**Drawbacks**	**References**
Heavy metal-contaminated soil	Municipal Waste Compost/Biosolids Compost (Wastewater sludge mixed with green wastes)	Increase of pH; reduction heavy metals (As, Pb, Cd, Cu, Zn); increase the total organic carbon and hydro soluble carbon content in soil	-	(Mora et al., 2005)
Areas of mine activities	Stabilized sewage sludge	Increase of pH, electrical conductivity, organic carbon and dehydrogenase activity	Sewage sludge may contain organic pollutants and potentially toxic elements	(Mingorance et al., 2014)
Tailings sites	Anaerobically digested sewage sludge or biosolids	Improve physical (increase gravimetric water retention and reduction of soil erosion), chemical (increase electrical conductivity, soil organic matter, total carbon and cation exchange capacity) and biological (increase total aerobic, total anaerobic, iron reducing, sulphate reducing and denitrifying microorganisms) properties	Heavy metal accumulation and N and P loading	(Gardner et al., 2010)
Abandoned opencast mining areas	Sewage sludge and nitrogen fertilizer	Increase total microorganism population, organic matter, total nitrogen, available nitrogen, phosphorus and potassium, soil biological fertility; prevent soil erosion (with vegetative cover)	Decreased C/N ratio	(Li et al., 2013)
Semiarid soil polluted with Cadmium	Dehydrated aerobic sewage sludge	Reducing inhibitory effects of Cd on biological parameters, increase biodiversity of soil microorganisms and their metabolic activity	-	(Moreno et al., 2002)

In some countries (e.g. in Australia, the USA, China, and some European countries) the recovery of biosolids as soil amendment/fertilizer in arable crops represent the most used (if not the main) disposal option (Mininni and Dentel, 2013). For instance, (Maria Cristina Collivignarelli et al., 2015), that carried out a regional study aimed at investigating the recovery of sewage sludge on agricultural land in Lombardy, reported that more than 90% of the Italian sewage sludge recovered in agriculture are applied to the soils of five regions (i.e. Lombardy, Emilia Romagna, Puglia, Tuscany and Veneto). However, at the moment, land application of biosolids is hampered by citizens because biosolids contain pathogens, inorganic and organic compounds that are or are perceived as dangerous for human health and for environment. Moreover, land application of biosolids releases odorous, volatile organic compounds (VOCs) (e.g., terpenes, alcohols, ketones, furans, sulphur-containing compounds, and amines) and ammonia (Maulini-Duran et al., 2013). Odours depend on initial substrate chemical composition, pH, moisture content, redox potential, atmospheric temperature, microbial activity, and physical and chemical properties of VOCs (Rosenfeld et al., 2001) and are released during their biodegradation. In many cases odours cause only discomfort to people living around fields where they are applied although (Lleó et al., 2013) report that exposure to high concentrations of odours produced by biosolids lead to toxicological effects (e.g. sensory irritation) and psychogenic effects.

In the scientific literature, no agreement can be found about the adverse effects caused by land application of biosolids; according with (Wang et al., 2008), they mainly refer to: (i) raising of the levels of persistent toxins in soil, vegetation and wild life, (ii) potentially slow and long-termed biodiversity-reduction through the fertilizing nutrient pollution operating on the vegetation, and (iii) greenhouse gas emissions (e.g. CH_4 and N_2O) (Figure 1).

Despite some heavy metals are considered as essential micronutrients for plant growth, high concentrations of these compounds could be toxic to food crops, domestic animals, and humans (Singh and Agrawal, 2008). It is also known that heavy metals are no biodegradable and their persistence in soil is much longer than any other reactive components of the terrestrial ecosystems. Therefore, the fate of heavy metals in post-biosolid-applied soil is of great importance with respect to interactions with the biological processes, their release and mobility, and transferability to the food chain.

In the soil, the bioavailability of heavy metals can increase and causes their excessive uptake by plants (which is correlated with extractable forms of metals rather than the total metal contents in soil) or leaching down. It must be highlighted that some plant species can protect the food chain by providing an effective barrier against the uptake of most heavy metals (Lu et al., 2012). The influence of biosolids on the availability of heavy metals and their effects on seed germination has been reported in literature (Islam et al., 2013; Walter et al., 2006). (Islam et al., 2013) investigated the effects due to the repeated application of biosolids to a silt-loam soils sited in Ohio (USA). Results revealed that the extractable fractions of Pb, As, Zn, and Cu were significantly higher at 0-15 cm soil depth. Consequently, the accumulated heavy metals may mobilize from the soils to groundwater and surface water bodies. Some authors

(Alvarenga et al., 2015; Cai et al., 2007; Hargreaves et al., 2008; Moretti et al., 2016) point out that the use of compost produced by the mixing of municipal solid wastes and sewage sludge can exceed, in some cases, the limits settled by *EU ECO Label for Soil Improvers* and by the *Proposed limit values for compost for the heavy metals,* especially for Ni, Pb and Zn (Cai et al., 2007; Hargreaves et al., 2008). Therefore, in order to immobilize heavy metals, alkaline stabilization of biosolids is identified as a better strategy, as well as the control of heavy metal concentration in biosolids before their reuse in agriculture.

Land application of biosolids may be also responsible for spreading human pathogens. The transmission of *pathogens* from application of sludge to humans, animals or plants is still a major concern on public health. Different physicochemical and biological parameters such as temperature, moisture content, oxygen, pH, sunlight, soil type, texture, and predation may influence the inactivation of pathogens in biosolids (Sidhu et al., 2001).

At present, specific limits for microbiological sludge quality or disinfection treatment requirements are not indicated in the Council Directive 86/278/EEC (CEU, 1986), which regulate the recovery of sewage sludge in the agricultural field. However, limits on microbial and organic compounds in the sewage sludge should be introduced; in fact, prevision concentration limits are reported in the *EU Working Document on sludge - 3rd draft* (EWA, 2000) and *EU Working document on sludge and biowaste* (DGEEC, 2010).

As concern the microbial parameters, the *Working Documents* (DGEEC, 2010; EWA, 2000) state that the treated sludge should fulfil the limits of *E.coli* < 500 CFU g^{-1} and the absence of *Salmonella spp.* in 50 g (wet weight). Additionally, sludge produced by conventional treatment shall, at least, achieve a 2-Log reduction in *E.coli* while any new sludge treatment process shall be initially validated through a 6-Log reduction of a test organism such as *Salmonella senftenberg W775*. As reported in (Mininni et al., 2015), the feasibility and reliability of these tests on sludge are still amply debated and seriously questioned because conventional indicators (e.g. *E.coli, faecal coliform bacteria, clostridia, somatic coliphages*, etc.) and/or pathogen index (*Salmonella*) are used as surrogate of pathogen presence for routine evaluation of treatment plant performances and sludge microbial quality. The problem is that viral pathogens show a differential persistence in the environment with respect to other microbial pathogens; therefore, the use of bacterial indicators is not providing reliable information of viruses reduction in sludge processing. The results of EU project ROUTES (CORDIS, 2014) proved that pathogens control should be focused on *Salmonellae, E.coli* and *somatic coliphages*, the latter ones resulted a very good indicator of enteric viruses. Since sludge can be applied on land once or twice a year (Fytili and Zabaniotou, 2008) pathogens can regrowth during the storage period and in the scientific literature there is an open discussion on the survival or regrowth of pathogens after sludge processing. Several studies have been focused on survival patterns and potential growth of inoculated organisms in sterile and non-sterile sludge;

only few studies have reported survival and regrowth of indigenous pathogens in sludge (Moce-Llivina et al., 2003; Pourcher et al., 2005).

In order to obtain a pathogen-free biosolids, thermal pretreatments such as thermophilic anaerobic digestion, pasteurization, and thermal hydrolysis can be used (NZWWA, 2003; USEPA, 1995; Wang et al., 2008). Performances of innovative sludge treatment trains are under study; for instance, (Levantesi et al., 2015) evaluated the performances of several advanced sludge treatment trains (i.e. thermophilic digestion integrated with thermal hydrolysis pretreatment; sonication before mesophilic/thermophilic digestion, and two sequential biological processes - mesophilic/thermophilic and anaerobic/aerobic digestion). As expected, they found that a better microbiological quality of sewage sludge was obtained with thermal treatments (e.g. thermal hydrolysis and thermophilic anaerobic digestion) even if the anaerobic/aerobic digestion process greatly contribute to the reduction of microbial load, allowing the achievement of the microbial quality levels proposed for the reuse of sludge in agriculture (only limited microbial load reduction was obtained by anaerobic digestion at 37 °C temperature and by mild sonication pretreatment).

As reported in (Manzetti and van der Spoel, 2015) sludge-based fertilization seems imprudent because of toxic compounds that may accumulate in the vegetation and then transferred to feeding herbivores and their predators. As a consequence, intoxication of foetuses, reduction in the reproductive potential as well as other long-term effects can compromise biodiversity and animal (human) proliferation. Conversely, several studies state that significant environmental or health risks connected to the use of biosolids on land have not been widely demonstrated (Clarke and Smith, 2011; M.C. Samolada and Zabaniotou, 2014). (Tejada et al., 2014) studied over two experimental seasons the effect of a biofertilizer obtained from sewage sludge on the yield and on the quality of maize crops (*Zea mays L.*). The results show that the application of sewage sludge had no effects on the soil, maize nutrition, grain quality or yield. In addition, they found that, in order to improve agricultural maize yields, quality and nutritional, sewage sludge should be applied as a foliar fertilizer instead of applying it to soil.

Over the past couple of decades, significant attention has been given to selected groups of persistent *organic contaminants (OCs)* in biosolids, including chlorinated dioxins/furans, polychlorinated biphenyls, and polycyclic hydrocarbons (Clarke and Smith, 2011). Most of these compounds do not affect human health when biosolids are recycled to farmland, possibly because of effective source control (Hundal et al., 2008). In EU about 143,000 chemicals are registered for industrial use; therefore, all of them could be potentially found in biosolids. Residual concentration of OCs depends, over their lipophilicity, on the initial concentration and on the extent of destruction during wastewater and sludge treatment. (Clarke and Smith, 2011) report that in the sewage sludge OCs account for few ng/kg to some percentage in the dry solids.

Research on organic contaminants in biosolids has been undertaken for over thirty years; recently (EWA, 2000) limit thresholds have been proposed for the so-called sum of halogenated organic compounds (AOX), linear alkylbenzene sulphonates (LAS), di(2-ethylhexyl)phthalate (DEHP), nonylphenole and nonylphenole ethoxylates (NP/NPE), polynuclear aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB), and polychlorinated dibenzo-p-dioxins and -furans (PCDD/F).

A number of emerging organic contaminants were identified in biosolids based on environmental persistence, human toxicity, and evidence of bio-accumulation in humans and in the environment. For instance, perfluorinated chemicals (PFOS, PFOA), polychlorinated alkanes (PCAs), polychlorinated naphthalenes (PCNs), organotins (OTs), polybrominated diphenyl ethers (PBDEs), triclosan (TCS), triclocarban (TCC), benzothiazoles, antibiotics and pharmaceuticals; synthetic musks, bisphenol A, quaternary ammonium compounds (QACs), steroids, phthalate acid esters (PAEs) and polydimethylsiloxanes (PDMSs) were recognized for priority attention because they can enter into living organisms via biosolids-amended soil (Brunetti et al., 2015; Clarke and Smith, 2011; Smith, 2009). Among those compounds, PFOS, PFOA and PCAs were identified for priority attention because they are environmentally persistent and potentially toxic or can be found in large concentrations in the sludge. Therefore, biosolids-amended soil can make theoretically possible to these compounds entering into human and ecological food-chains. However, there is a growing body of evidence demonstrating that the majority of the studied compounds do not endanger human health when they are recycled to farmland (Clarke and Smith, 2011; Smith, 2009).

Recently, (Braguglia et al., 2015) investigated the performances of different enhanced sludge stabilization processes on a broad class of conventional (EOX, LAS, NPEs, PCBs, PAHs, and phthalates) and emerging organic micropollutants contained in digested sludge. Processes studied were: (i) thermophilic digestion integrated with thermal hydrolysis pretreatment, (ii) sonication before mesophilic/thermophilic digestion, and (iii) sequential anaerobic/aerobic digestion. Results indicated that the concentrations of the conventional organic pollutants in the feed just in few cases exceed the recommended thresholds set in the (DGEEC, 2010). Removals of conventional and emerging organic pollutants were greatly enhanced by performing double-stage digestion (sonication before mesophilic/thermophilic digestion and sequential anaerobic/aerobic digestion treatment) if compared to a single-stage process (such as thermal hydrolysis pretreatment). As concerns the toxicity reduction, the authors found similar results.

Finally, as reported in several studies (Aguilar-Chávez et al., 2012; Majumder et al., 2014; Maulini-Duran et al., 2013; Nkoa, 2014) biosolids contribute to greenhouse gas (GHG) emissions in different stages, mainly stockpiles and their land application. For instance, (Majumder et al., 2014) studied the direct emission of GHG generated from biosolid stockpiles in Melbourne (Australia) and found that the youngest biosolids (< 1 year) released higher amounts of methane (CH_4) and nitrous oxide (N_2O). In comparison, stockpiles aged between 1 and 3 years emitted higher overall

GHGs compared with the oldest stockpiles. Studies revealed that GHG emissions were dominated by CO_2 and N_2O while CH_4 is emitted in low concentration (accounted for less than 2%) and generally its contribution can be considered negligible (Majumder et al., 2014). In order to minimize GHG emissions from biosolids applications, some actions such as selecting remote sites, minimizing the length of time for storage of biosolids can be undertaken.

5. Recovery of sludge in engineering applications

In this section, the use of sewage sludge in the mixture of construction and building materials is reviewed.
Sludge recovery for producing e.g. bricks, lightweight artificial aggregates, and cement-like materials is considered worldwide a win - win strategy because it converts the wastes into useful materials by the concomitant reduction of disposal issues (Ukwatta et al., 2015). Moreover, reusing, reprocessing, or recycling materials reduces extraction of raw resources (Calkins, 2009).

Construction industry is a suitable technological activity sector to employ solid wastes, due to the large amount of raw materials and final products used (Martínez-García et al., 2012; Ukwatta et al., 2015). In fact, as reported in (Calkins, 2009), each year more than three billion metric tons of raw materials are used to manufacture construction materials and products worldwide. In addition, the construction industry has to tackle the problem of the depletion of natural materials such as pumice, scoria, crushed stones, and clay. In some countries, the exploitation of raw material is becoming severely regulated; for instance, in order to protect the clay resource and the environment, China have started to limit the use of bricks made from clay (Chen et al., 2011). Following, the prospective benefits deriving from the use of sewage sludge in the construction and building materials as well as the mechanical proprieties of the obtained materials are reported (see also figure 1).

5.1 Road construction

The feasibility of sludge recovery in the field of road engineering is reported in several works (de Figueirêdo Lopes Lucena et al., 2014; Kanari et al., 2016). The studies were focused on performances assessment of different kinds of sludge that are mainly employed in the road base layer or as a fill material in road embankments as substituted of raw materials.

The growth of plants on road embankments is of great importance for their management. Plants growing on embankments help decrease pollution, provide a desirable touch of natural beauty, and protect the roadbed. Generally, embankment soil is basically selected according to its resistance characteristics; therefore, in some cases, its agronomic characteristics are very limited (e.g. immature, bad soil structure, and low nutrient content) (De Oña and Osorio, 2006; Pengcheng et al., 2008). The growth and development of grasses and counteracts is important because they protect the roadbed from erosion (de Figueirêdo Lopes Lucena et al., 2014; De Oña et al., 2011, 2009; Pengcheng et al., 2008). Several studies were carried out for assessing the viability of sewage sludge utilization (de Figueirêdo Lopes Lucena et al., 2014; De Oña et al., 2011, 2009; Pengcheng et al., 2008).

(De Oña and Osorio, 2006) and (Pengcheng et al., 2008) studied the effects on the growth and development of plants due to the sewage sludge utilization. Both authors investigated the addition of sewage sludge compost (SSC) to a natural soil. Results show that the use of SSC improved the chemical-physical properties of the soil, increased the growth of the plants (they used the perennial ryegrass), reduced the volume and the total mass flux of sediments in runoff. Moreover, (De Oña et al., 2009) report that sludge–compost mixtures worked better than the use of sludge only or compost only.

As concern the potential threat related to the use of sewage sludge in the road construction field, the available researches suggest that sewage sludge, if treated properly and managed in accordance with existing regulations and standards, is safe for the environment and human health (Arulrajah et al., 2011; De Oña and Osorio, 2006; Pengcheng et al., 2008). (Arulrajah et al., 2013), for instance, indicated that heavy metals, dichloro diphenyl trichloroethane (DDT) and organochlorine pesticides concentration along with pathogens (bacteria, viruses, or parasites) were within acceptable limits for usage in geotechnical applications.

Engineering properties of sewage sludge have been studied in recent years in several countries such as UK, Hong Kong, USA, South Korea, Turkey, Spain and Singapore (Arulrajah et al., 2011). In literature, some tests such as California Bearing Ratio (CBR), unconfined compressive strength, indirect-tensile strength, resilient modulus, and deterioration tests were conducted for investigating mechanical properties of sewage sludge (pure or blended) (Arulrajah et al., 2013, 2011; de Figueirêdo Lopes Lucena et al., 2014).

(Arulrajah et al., 2013) report that biosolids tested have similar properties to soil, such as moisture content, cation exchange capacity, and moisture retention as well as geotechnical engineering properties (e.g. plastic behaviour, acceptable shear strength parameters and compaction ability). However, several research studies show that sewage sludge is generally associated with high compressibility (Disfani et al., 2009), high rates of creep, and possible unsatisfactory strength characteristics (Suthagaran et al., 2008), which increases the risk of excessive settlements in case of their application as load bearing media (Santagata et al., 2008). As reported in some researches, bearing capacity can be improved submitting biosolids, before the use, to a stabilization process even if best results can be obtained blending biosolids with additives such as cement, lime, and emulsion (de Figueirêdo Lopes Lucena et al., 2014; Disfani et al., 2009; Suthagaran et al., 2008). For instance, (de Figueirêdo Lopes Lucena et al., 2014) investigated the possibility of using 10 wt% of sewage sludge in pavement base layers, adding, in different percentages (2, 4, 6, and 8% by weight), three additives (i.e. cement, lime, and emulsion). Results indicated that the CBR gains when using lime and cement as additives and decreases when using emulsion. In particular, the addition of 8 wt% cement to the mixture of soil sewage sludge supplied the highest increments of resistance. Similar results were also obtained by (Suthagaran et al., 2010, 2008).

5.2 Bricks and ceramic products

Bricks have been a major construction and building material for a long time (the first applications of dried-clay bricks were in the 8000 BC - Before Christ) (Zhang, 2013).

Conventional bricks are produced from clay and shale with high temperature kiln firing or from ordinary Portland cement (OPC) concrete (Calkins, 2009; Zhang, 2013).

Brick-making sector is characterized by low energy efficiency (CCAC, 2015); for instance, (Calkins, 2009) report that clay bricks require from 150% to 400% more energy to produce than concrete paving bricks. Low technological levels are highly related with pollutant air emissions; in fact, brick production contributes with greenhouse gases (GHG) and black carbon (BC) emissions, with a significant impact on human health and climate change. Recent studies show that the implementation of more efficient technologies can reduce the pollutant emissions from 10 to 50%, depending on the process, scale and fuel used (CCAC, 2015). Moreover, bricks-making sector is characterized by an intensive quarry activity; in the 2014, only in the USA, about 10 million tons of common clay have been mined (Jewell and Kimball, 2015). In order to reduce the impacts related to quarry activities, saving the costs and for a sustainable development, (The Brick Industry Association, 2015) report that almost 50% of manufacturers incorporate some kind of waste into their bricks.

Many researchers evaluated the use of a wide variety of waste materials, including, for instance: fly and bottom ash (Arıöz et al., 2010; Chen et al., 2012), fly ash from coal-fired generators (Freidin, 2007), mine tailings (Ahmari and Zhang, 2012), cigarette butts (Abdul Kadir et al., 2009), and rice husk ash (Hegazy et al., 2012a, 2012b).

The incorporation of sewage sludge into the bricks – possibly blended with other materials (e.g. fly ash, circulating fluidized bed combustion bottom ash, agricultural wastes, forest wastes, etc.) – was proposed and researched since the eighties of the last century (e.g. (Alleman and Berman, 1984)).

Researches are mainly focused on investigating the effects of the addition of sewage sludge in different percentages; typically, they are blended from 2% up to 50% by weight (Ingunza D. et al., 2011; Kadir and Mohajerani, 2011; Ukwatta et al., 2015) although other authors (for instance (Weng et al., 2003) and (Tay et al., 2004)) investigated also the proprieties of bricks made with 100% of sewage sludge.

Tests conducted by many authors indicated that the proportion of sludge in the mixture and the firing temperature are the two key factors affecting the brick quality (Weng et al., 2003). In general, the addition of sludge in proportions of 2 up to 20 wt% does not induce significant changes in the relevant functional characteristics of bricks (Martínez-García et al., 2012); on the contrary, a higher amount of sludge in the mixture could compromise their characteristics (Ingunza D. et al., 2011; Liew et al., 2004a; Tay et al., 2004; Weng et al., 2003). For instance, (Liew et al., 2004a, 2004b) reported that bricks with a sludge content of up to 40 wt% were capable of meeting the relevant technical standards although bricks with more than 30 wt% sludge addition are not recommended (they are brittle and easily broken even when handled gently). In addition, (Ingunza D. et al., 2011) found that bricks with 35 wt% of sludge were reduced in some dimensions between 1 mm to 7 mm.

During the firing process *brick mass* can significantly decrease due to the sewage sludge organic matter reduction; for instance, (Liew et al., 2004a) and (Ukwatta et al., 2015) report that the weight loss on ignition increase according to the percentage of sludge within the bricks. (Martínez-García et al., 2012) report that the decomposition of organic matter occurred between 200 and 550 °C. Authors found that the first exothermic peak (200 - 400 °C) is associated with biodegradable materials, undigested organics, and dead bacteria, as well as the emission of semivolatile compounds.

The degrees of *firmness and compaction* of bricks, as measured by their water absorption characteristics, vary considerably depending on factors such as the type of clay and methods of production used. Many authors report that the water absorption of the bricks increases with increased sludge addition and therefore leads to decreased resistance to weathering (Ingunza D. et al., 2011; Kadir and Mohajerani, 2011; Martínez-García et al., 2012; Ukwatta et al., 2015). For instance, (Ingunza D. et al., 2011) used 25 wt% of sludge in the mixture and found that bricks absorbing capability increased to an average of 160% more than control brick. (Liew et al., 2004a) and (Jordán et al., 2005) found that *water absorption* increase with sludge percentage with a linear relation.

The firing temperature and the proportion of sludge in the mixture are the parameters affecting the degree of *shrinkage* (Weng et al., 2003). Since the swellability and the organic content of the sludge are much higher than those of clay, the addition of sludge in the mixture should increase the degree of firing shrinkage although in literature results are not in agreement. For instance, (Martínez-García et al., 2012) and (Ukwatta et al., 2015) found that the shrinkage growth with the increase of sewage sludge in the mixture, while, the opposite results are reported in (Liew et al., 2004a, 2004b), (Jordán et al., 2005) and (Monteiro et al., 2008).

The *compressive strength* is the most important requirement for assuring the engineering quality of a building material. Authors that investigated this parameter agreed that the strength is greatly dependent on the amount of sludge in the brick: higher amounts of sewage sludge in the mixture involve lower strength (Ingunza D. et al., 2011; Martínez-García et al., 2012; Ukwatta et al., 2015). For instance, (Ukwatta et al., 2015) found that the addition of 25 wt% of biosolids in the mixture implies a reduction of more than 50% of the compressive strength of the brick samples (strength passed from 36.1 MPa (control) to 16.2 MPa for bricks made with 25% of biosolids). This finding was confirmed by (Ingunza

D. et al., 2011) even if they found that also a slight addition of sludge in the mixture (5 wt%) can significantly affect the compressive strength performance of bricks (their lost up to 70% of maximum strength in the bricks manufactured with 20 wt% of sewage sludge).

(Weng et al., 2003) also investigated the effect of the *firing temperature* on the bricks strength. Results indicate that the strength is greatly dependent on the amount of sludge in the bricks. In fact, bricks with up to 10 wt% of sludge in the mixture and cooked at 1000 °C reached strength values close to the standard clay bricks (roughly 20 MPa) although it decreased about 50% cooking the bricks at 880 °C.

In addition, it was found that *sewage sludge moisture* could affect the mechanical proprieties of final products; for instance, (Weng et al., 2003) recommended to use sewage sludge with 24% water content, while (Yagüe et al., 2002) report that the addition of 2 wt% dry pulverized sludge can significantly increase the compressive strength, decrease the porosity and water absorption.

(Martínez-García et al., 2012) also investigated the *freezing resistance*, which is defined by the decrease of samples compressive strength before and after undergoing 25 ice-defrosting cycles. After 25 ice-defrosting cycles, they proceeded to the eyepiece inspection of the specimens and no cleavage, fissure or scalping have been encountered in samples with sludge content lower than 15 wt%; superficial deterioration has been observed in the case of samples with higher sludge content.

Finally, (Ukwatta et al., 2015) measured the *thermal conductivity* of bricks produced with different amount of biosolids. Results show that thermal conductivity decreased from 1.08 (control) to 0.81 W m^{-1} K^{-1} in the bricks casted with 25 wt% of biosolids in the mixture with positive implication in terms of energy savings.

Despite many authors have demonstrated the feasibility, by a mechanical point of view, of the utilization of sewage sludge for brick construction, some sectors of public opinion are against to put in practice that process due to the sanitary safety of buildings built with these materials.

The environmental behaviour of construction product is assessed by the study of the properties that influence its environmental sustainability, such as *leaching behaviour*. As there is no harmonization in the tests and components to be studied to determine the environmental performance of the waste-based ceramic products so far, the leaching tests selected in each case are different (Pacheco-Torgal et al., 2015). The most used tests are the Toxicity Characteristic Leaching Procedure (TCLP) (Liew et al., 2004a; Martínez-García et al., 2012; Weng et al., 2003) and the diffusion leaching test – NEN 7345 (Cusidó and Cremades, 2012; Cusidó and Soriano, 2011). In general, authors agreed that heavy metals are the main compounds which can be found in the leachates from bricks made with sewage sludge; authors highlight that heavy metals are originally present in the sludge or in the clay and the leaching from the bricks is very low (Liew et al., 2004a; Martínez-García et al., 2012; Weng et al., 2003). (Weng et al., 2003) have recorded that chromium and zinc were leached in greater amount with respect to the other heavy metals, although the concentrations were much lower than those of the Taiwan-EPA regulated TCLP limits; moreover, organic compounds from sewage sludge do not appear in leachates. (Cusidó and Cremades, 2012) investigated the potential health risks related to people who live in houses built with materials made from sewage sludge. Tests were conducted according with (ESA PSS-01-729, 1991) and (ESA PSS-01-702, 1994). By means of these tests it is possible to evaluate the gases (i.e outgassing and offgassing) and particles emitted by the bricks in a simulated time frame equivalent of 10 years. Also in this case, tests show that there are no environmental restrictions on the use of clay bricks made with sewage sludge.

Finally, *the visual appearance* of bricks can be influenced by many factors such as the firing temperature and the amount of sewage sludge in the mixture although authors do not found the same results. (Liew et al., 2004b) and (Cusidó and Cremades, 2012) found that a high amount of sludge addition in mixture has a pronounced effect on the pore structure of the amended clay bricks, involving uneven and rather poor surface textures. Moreover, (Kadir and Mohajerani, 2011) report that the firing process can cause black coring to the final product. Authors concluded that sludge bricks might not be suitable as facing bricks due to their poor surface texture and finishing, unless wall plasters (cladding or rendering) are applied. On the contrary, (Ingunza D. et al., 2011) found that there is no sign of alteration in colour or odour in the bricks made with up to 20 wt% of sewage sludge in the mixture.

5.3 Lightweight aggregates

Nowadays, in the construction sector there is a great interest for the use of natural materials and/or aggregates that undergo thermal expansion under controlled conditions for the production of *lightweight aggregates (LWAs)*. The perlite and some lamellar minerals (i.e. vermiculite, clay, schist, shale, slate) are the most used raw materials for the thermal synthesis (Kanari et al., 2016). An excessive exploitation of these non-renewable natural resources will lead to their depletion in the future. Thus, in order to preserve the reserves in granulates, the use of residues derived from waste industry could represent an interesting solution.

Several studies (Chiang et al., 2009; Franus et al., 2016; González-Corrochano et al., 2016; Mun, 2007; Tuan et al., 2013; Wang et al., 2009) investigated the effects of using both biosolids and water treatment sludge (WTS) in the production of LWAs. Most previous studies have focused that the use of dewatered sewage sludge involves the production of porous and loose aggregates due to high organic matter and water content in sewage sludge. Thus, generally, no greater than 30% sewage sludge should be used. In practical, in order to improve the performance of manufactured LWAs, sewage sludge could be mixed with suitable materials such as coal ash (Wang et al., 2009), inorganic waste (Tuan et al., 2013), organic waste (Chiang et al., 2009) and clay (Tay et al., 1991).

Regarding the application of biosolids as a substitute of sand/stone, LWAs need to satisfying the strength requirement of ASTM C330 and ACI 318 for structural lightweight concrete, which requiring a minimum 28-day compressive strength of 17.2 MPa (Tuan et al., 2013). *Compressive strength* reported by several research varies between 24 and 60 MPa, these results complies the value limit. This parameter is affected by (i) temperature and (ii) material mixed with sewage sludge. (Wang et al., 2009) and (Chiang et al., 2009) showed an increase (more than double) of compressive strength when sintering temperature goes up to 1050 °C to 1100 °C. (Chiang et al., 2009) also investigated the effect of organic residues mixed with sewage sludge: the results showed an increase of compressive strength with a decrease of rice husk added. This result was not confirmed by (Wang et al., 2009), that did not report any linear correlation between these parameters.

As concern the *bulk density***,** different authors (Huang and Wang, 2013; Tuan et al., 2013) measured different values (0.5-1.5 g cm^{-3}), which is mainly related to sintering temperature and percentages of material mixed with sewage sludge such as compressive strength. As reported in (Tuan et al., 2013) an increasing of temperature, for instance from 850 °C to 1100 °C, and percentages of waste glass powder mixed with sewage sludge, from 30% to 50%, involved a reduction of bulk density about 20-30%. In opposite, with the same sintering temperature, but with a 10% of waste glass powder mixed with sewage sludge, they showed an increase (of 10%) of bulk density.

Several authors also investigated the *water absorption*. (Huang and Wang, 2013) observed that the water absorption rates of the LWAs ranging from 0.5% to 15%. The increase of different materials/residues, such as clay (Tay et al., 1991), rice husk (Chiang et al., 2009) and coal ash (Wang et al., 2009), mixed with sewage sludge involved a growth of water adsorption. However, (Tuan et al., 2013) highlighted that the water absorption decrease when increase the amount of waste glass powder mixed with sewage sludge.

The most important parameter that affects the water absorption of LWAs is the sintering temperature. Generally, as reported by (Tuan et al., 2013), the water absorption of sintered samples decreased when the heating temperature increased. Moreover, higher sintering temperatures are advantageous for the stabilization of heavy metals, that can be stabilized in LWAs, preventing their release and secondary pollution of the environment (Xu et al., 2013).

5.4 Cement

Concrete is one of the most commonly used construction material in the world (Khatib, 2016). Portland cement, the primary constituent of concrete, is produced and used in large quantities: for instance, about 237 million tons only in the European Union. It is well known that the production of OPC (Ordinary Portland Cement) is highly energy intensive (the production of 1 ton of OPC consumes approximately from 2.6 to 6.2 GJ of energy) and releases significant amount of greenhouse gases (the production of 1 kg of OPC generates approximately 0.8–0.9 kg CO_2 emissions) (Calkins, 2009; Zhang, 2013). The calcination of carbonate rocks during cement production is the more impacting stage of the cement industry: this phase accounted for about 5% of global CO_2 emissions from all industrial process and fossil-fuel combustion in 2013 (Xi et al., 2016). This negative record is mainly due to the use of coal as primary energy source for cement production (followed by petroleum coke and purchased electricity) and to the chemical conversion from the calcination of lime-stone and other carbonate-containing feedstocks (Cagiao et al., 2011; Chen et al., 2010).

Strategies that can be used for minimizing the environmental impacts of cement are twofold: (i) the reduction of the cement use in a concrete mixture, and (ii) the cement replacement with appropriate alternative raw materials and fuels.

Reduction in cement use in a concrete mixture is most easily achieved through the replacement of OPC with other pozzolanic or hydraulic materials. A research carried out by (Aïtcin, 2000) had shown that the use of less cement is possible (by specifying a 56-day full-strength requirement instead of the traditional 28-day full-strength requirement), in addition more durable structure can also be obtained. The most common *supplementary cementitious materials* (SCMs) are industrial by-products used in the concrete mixture; these include, for instance (Khatib, 2016; Strigáč, 2015): ground-granulated blast-furnace slag, silica fume, metallurgical slags, siliceous and calcareous fly ashes, circulating fluidized bed combustion fly and bottom ashes, spent foundry sand, construction and demolition waste, chemical gypsum and sewage sludge. As reported in many researches, municipal sewage sludge can be used as *alternative fuel* (Lin et al., 2012; M. C. Samolada and Zabaniotou, 2014; Yan et al., 2014) and as *substitute of raw materials* (Barrera-Díaz et al., 2011; Liu et al., 2015; Valderrama et al., 2013; Yen et al., 2011). Among the sludge derived from WWTPs, dried sewage sludge is the most investigated for energy and raw materials recovery in the cement kilns factories (Husillos Rodríguez et al., 2012; Rulkens, 2008; M. C. Samolada and Zabaniotou, 2014; Werle and Wilk, 2010). Moreover, digested sludge (Tay et al., 2002), waterworks sludge (Chen et al., 2010), and dried industrial sludge (Arsenovic et al., 2012) have been investigated.

The use of sewage sludge in the cement production is influenced by many factors although the co-processing of sewage sludge in cement kilns has yet been widely employed at the full-scale plants in the United States, Europe, Japan and other developed countries (Lv et al., 2016; Rahman et al., 2015).

According to (Stasta et al., 2006), sludge can be used in the cement kilns if comply, at least, with the following characteristic parameters: (i) maximum moisture content of 20%, (ii) low heat value (LHV) of 9 MJ kg^{-1} and (iii) granulometry between 0 and 5 mm.

Sludge produced in WWTPs contains useful compounds that can be used for the production of OPC; for instance, sewage sludge contain CaO, SiO_2, Al_2O_3 and Fe_2O_3 that represent, since as a first approximation, the four major oxides of Portland cement clinker (Pacheco-Torgal et al., 2013; Valderrama et al., 2013; Yen et al., 2011). Other useful compounds that can be find in the sludge and that could affect the burning process (clinkering, cooling, and emission) of the Portland cement are *chlorides* (typical concentration of those compounds are reported in Table 1) and *phosphate*. Chlorides, as reported in (Kwon et al., 2005) and (Maki, 2006), increase the burnability of the raw meal and allows higher contents of alite (tricalcium silicate, 3CaO SiO_2, called as C_3S) at the same clinkering temperature. Moreover, chlorides have a great capacity for reducing the viscosity of the liquid phase and can improve the solubility of CaO (CaO is highly soluble in liquid phases rich in halogen).

Phosphate in Portland cement should range between 0.3 to 0.5 wt%, typically it is in the order of 0.2%. The effects of phosphate on the characteristics of cement clinkers made with sewage sludge were investigated by many authors (Fukuda et al., 2010; Kwon et al., 2005; Lin et al., 2009; Moudilou et al., 2007). In laboratory experiments performed by (Fukuda et al., 2010) and (Pacheco-Torgal et al., 2013), it was shown that the addition of a small amount of P_2O_5 suppress the 'dusting effect' due to the transformation of β-C_2S to γ-C_2S. Authors agreed that sludge incorporation into cement raw meal was effectively limited by the phosphate content which, up to 0.7%, began to increase belite (dicalcium silicate, 2CaO SiO_2, called as C_2S) formation at the expense of alite causing increased setting times and lower strength development in pastes. Moreover, also the Sulfur (S^{6+}) content can influence the characteristics of cement; in fact, (Pacheco-Torgal et al., 2013) report that SO_3 and P_2O_5 decrease both the viscosity and surface tension of the liquid as well as the polymorphic form of C_3S. In addition, (Naamane et al., 2016) show that the high amounts of P_2O_5 and SO_3 in sewage sludge calcined in temperature range 700–800 °C increase water demand and setting time compared to the control mortar. The addition of SO_3 or SO_3 + HPO_4^{3-} simultaneously reduces the burnability, whereas it is improved with the addition of SO_3 + HPO_4^{3-} and F^- (Maki, 2006). Finally, alkali metal oxides (Na_2O and K_2O) increase the viscosity and decrease the surface tension of the liquid phase (Pacheco-Torgal et al., 2013).

As reported in (Naamane et al., 2016) the presence of absorbed water and organic matter in sewage sludge calcined in temperature range 300–500 °C prolongs strongly the setting time and affects negatively the compressive strength and the hydration degree of mortars.

As shown in Table 1, *heavy metals* can be found in the sludge. Many authors (Espinosa and Tenório, 2000; Gineys et al., 2011; Kakali et al., 1990; Murat and Sorrentino, 1996; Stephan et al., 1999a, 1999b) investigated the effects of heavy metals on cement properties. For instance, (Gineys et al., 2011) explored the maximum amount of Cu, Ni, Sn, and Zn that could be incorporated in a laboratory clinker and found the following threshold limits: 0.35% of Cu, 0.5% of Ni, 1% of Sn and 0.7% for Zn. (Murat and Sorrentino, 1996) and (Espinosa and Tenório, 2000) studied the effects on cement properties when adding a sludge containing Cr as the predominant heavy metal. Authors concluded that the largest amount of Cr was trapped in Portland cement. All authors concluded that Cr, Ni, and Zn in the sewage sludge had no impact on cement mortar strength or initial setting time or hydration of cements because are typically lower than threshold limits.

The *amount of sludge that can be added as raw material* substitute can range from 5 to 15 wt% (Husillos Rodríguez et al., 2012; Johnson et al., 2014; Lin et al., 2012). Different authors studied the effects of dried sewage sludge as an additive on cement property in the process of clinker burning. Authors refer that, due to the organic content of the sludge, in order to avoid undesirable changes in the mechanical and rheological properties of pastes and mortars, sludge replacement may not exceed a replacement rates greater than 10%.

Sewage sludge can be also extensively used in cement manufacturing *as a cheap alternative energy resource* with substantial energy and environmental savings (its CO_2 emissions are lower than coal) (Husillos Rodríguez et al., 2012; Liu et al., 2015; Rovira et al., 2011). Moreover, co-combustion of sewage sludge in cement kilns represents an advantage for a low investment cost and rapid implementation (Zabaniotou and Theofilou, 2008) usually, there are no additional investment costs for off-gas cleaning (Stasta et al., 2006). (Wang et al., 2008) refer that dewatered biosolids can be utilized instead of dried sludge; approximately 5 wt% may be co-fired together with coal without compromising the temperature of the combustion process.

The effects on the air emissions due to the co-processing of sewage sludge in cement kiln are complicated. For instance, (Cao et al., 2013), (Liu et al., 2015) and (Fang et al., 2015) showed that sewage sludge can be used as a reducing agent for NO_x removal. (Fang et al., 2015) investigated, especially, the influences of sludge feed rate, feed point, feed method, and air-staged combustion on NO_x removal. Results indicate that the use of sludge as a secondary fuel is conducive to NO_X reduction, which depends primarily on the feed rate and feed point.

Conversely, sewage sludge can also make the pollutants more complex, even cause the emission of unconventional air pollutants, such as PAHs, dioxins and heavy metals (Lv et al., 2016; Rovira et al., 2014, 2011). For instance, when sewage sludge is co-processed in cement kiln, PAHs emission shows a trend of increase although its emission is small (Conesa et al., 2011; Gálvez et al., 2007).

Some authors mainly focused on investigating the effects on human health risks derived from the exposure to PCDD/Fs and metals (Rovira et al., 2014, 2011) emitted by a cement kiln that co-process sewage sludge. (Rovira et al., 2011) found that PCDD/Fs emission slightly increases when sewage sludge are co-processed though they were within the ranges considered acceptable by international regulatory organisms (Rovira et al., 2011). As concern heavy metals, (Stasta et al., 2006) and (Rulkens, 2008) agreed that they are immobilized within the cement.

6. Other recovery options

6.1 Adsorbent materials

An alternative route of sewage sludge recovery is the conversion into adsorbent material with sustainable methods to allow its reuse in water treatment applications (Wu et al., 2015; Xu et al., 2015). The first to recognize sewage sludge's potential as a feedstock for producing activated carbon was (Kemmer et al., 1971), since then different study analysing the production of adsorbent from sewage sludge by its carbonisation (Smith et al., 2009).

Adsorbent material is obtained from conversion of sewage sludge via pyrolysis, which allow to achieve, therefore subjecting to an activation process, the production of char, a low cost adsorbent with good adsorption properties in water treatment applications (Hadi et al., 2015; Kimbell et al., 2018; Liu et al., 2010; Rio et al., 2006; Smith et al., 2009).

Numerous methods of activating carbons are available, but it's possible grouped them in two categories: physical activation and chemical activation (Smith et al., 2009).

Physical activation of sewage sludge is commonly carried out with carbon dioxide (Jindarom et al., 2007; Marques et al., 2011; Ros et al., 2006), steam (Li et al., 2011; Rio et al., 2006; Smith et al., 2012) or air (Monsalvo et al., 2011) and prescribes two steps: carbonisation and activation (Alvarez et al., 2016). Carbonisation allows breaking down the cross-linkage between carbon atoms (Alvarez et al., 2016) in order to increase the Brunauer–Emmelt–Teller (BET) surface area of the resulting char. The main parameters that influence this process are heating rate and dwell time: different results are present in literature (Jeyaseelan and Qing, 1996; Méndez et al., 2009; Seredych and Bandosz, 2007; Yilmaz et al., 2011; Zhai et al., 2004) to obtained a good carbonisation other factors are important: mesoporosity, macroporosity and feedstock type, which were reported by (Rio et al., 2005, 2004), (Weng et al., 2001) and (Ding et al., 2012). The transformation of sewage sludge in char is completed by activation with gas at high temperature (800-1200 °C) for further development of the sludge-based adsorbent's (SBA) porosity (Alvarez et al., 2016). Lots of activation agents are reported in literature, including N_2, CO_2, steam, O_2/Air, etc.; in general, steam (Alvarez et al., 2016) and CO_2 (Alvarez et al., 2016; Smith et al., 2009) are the most commonly used.

Another possibility is chemical activation, which depends on temperature, activator type and concentration and binder addition. There are a wide variety of activators with different activation temperature, but the most common used include KOH, NaOH, $ZnCl_2$ and H_3PO_4 (Alvarez et al., 2016). In particular, KOH was proved to be an effective activator in producing SBAs with high BET surface areas when is obtained through carbonisation and activation (Alvarez et al., 2016; Smith et al., 2009).

Results of conversion of sewage sludge to adsorbent depend on different treatments (physical or chemical) and parameters (temperature, time, acid washing). Generally surface areas of char ranges from 100 to 2000 $m^2 g^{-1}$, where the best results are obtained with chemical activation. In fact, use of KOH gives the opportunity to reach BET surface areas between 1000 and 1900 $m^2 g^{-1}$ (Lillo-Ródenas et al., 2008; Ros et al., 2006; Shen et al., 2008, 2006; Smith et al., 2009), but high value can be achieved from activation with NaOH, 1224 $m^2 g^{-1}$ (Ros et al., 2006), or ZnCl2, 700 $m^2 g^{-1}$ (Chen et al., 2002; Tsai et al., 2008), too.

Bet surface areas obtained with physical activation vary from 100 to 500 m^2 g^{-1} (Bandosz and Block, 2006) due to temperature, time and acid washing. Different temperature and time are investigated by different authors (Seredych and Bandosz, 2007; Zhai et al., 2004) as well as acid washing with HCL, which dissolve inorganic content with a consequence increase of surface of char (Ros et al., 2007).

The adsorbent material obtained from sewage sludge may be used for different applications, the most applied is the adsorption of volatile organic compounds (VOCs) (Anfruns et al., 2011; Benintendi, 2016): removal of NO_x (Pietrzak and Bandosz, 2008, 2007) and H_2S (Bandosz and Block, 2006; Sioukri and Bandosz, 2005) are typical examples. The adsorbent could be used for adsorption of dyes, phenolic compounds and antibiotics too (Kimbell et al., 2018). The adsorption of anionic and cationic dyes is reported in different articles (Bandosz and Block, 2006; Rozada et al., 2003) and the adsorbate for assessing the dye uptake of carbons is methylene blue (Smith et al., 2009). The methylene blue adsorption capacity varies from 200 mg g^{-1} to 500 mg g^{-1} (Gómez-Pacheco et al., 2012), when was obtained by a NaOH activated SBAs. As regards the adsorption by carbonaceous adsorbents of phenol/phenolic compounds and antibiotics are describes by (Dąbrowski et al., 2005) and (Ding et al., 2012), respectively. Another important application is the adsorption of heavy metal: cadmium (Gutiérrez-Segura et al., 2012), hexavalent chromium (Agrafioti et al., 2014; Deng et al., 2010), mercury (Bandosz and Block, 2006) are typical examples.

The two most significant factors for the sewage sludge-based adsorbents to evaluate their economically feasible application are adsorption capacity and cost. The cost of SBAs depends on various factors, including local availability, nature of sewage sludge, processing required, preparation conditions and both recycle and lifetime issues (Xu et al., 2015). The production of SBAs costs approximately 0.1-0.2 US \$ kg^{-1}, which is cheaper than commercial activated carbon (2.0-2.2 US \$ kg^{-1}) (Ahmaruzzaman, 2011; Lin and Juang, 2009), in addition to a good capacity of adsorption: for examples high methylene blue adsorption capacity (260 mg g^{-1}) is connected with a low cost (365 US \$ t^{-1}) (Xu et al., 2015).

6.2 Phosphorous recovery

Sewage sludge has a high phosphorus content (approximately 8% w/w), making it a potential source of nutrients. Phosphorous recovery process from biological sludge is composed in relation to the different technologies and different characteristics of organic matter used (sludge liquor, digested or non-digested sludge). Direct extraction of P from sewage sludge allows to reduce the high energy associated with ashing of sewage sludge, that represents a commonly practiced in most European countries (Shiba and Ntuli, 2017). Recovery from sewage sludge requires a prior hydrolysis, disintegration and dissolution, while from liquid phase the principal treatments concerns the precipitation or crystallization (Blöcher et al., 2012). P-recovery through precipitation can be subdivided in different group:

precipitation in the sludge with or without prior leaching, adsorption to a carrier and pellet formation (Sartorius et al., 2011). These techniques are based on minerals precipitation in the form of struvite, hydroxyapatite or calcium phosphate. The most important advantage is the ability to obtain high-quality phosphoric minerals and the use of sludge for direct applications in agriculture (Cieślik and Konieczka, 2017). Furthermore, precipitation of struvite allows to improve the compost quality (if composting is the final recovery of sludge) through conservation of nitrogen: it is shown a gradually increased and stabilized concentration of NH_4 when struvite precipitation is applied in composting process (Kataki et al., 2016). Also for this reason, precipitation is the major process adopted for sewage sludge P-recovery (Kataki et al., 2016). (Shiba and Ntuli, 2017), by means of acid leaching followed by ion exchange and precipitation using magnesium hydroxide and ammonium hydroxide, shown a technique for recovering the P nutrient (about 82% of P was extracted as calcium phosphates and aluminium phosphates).

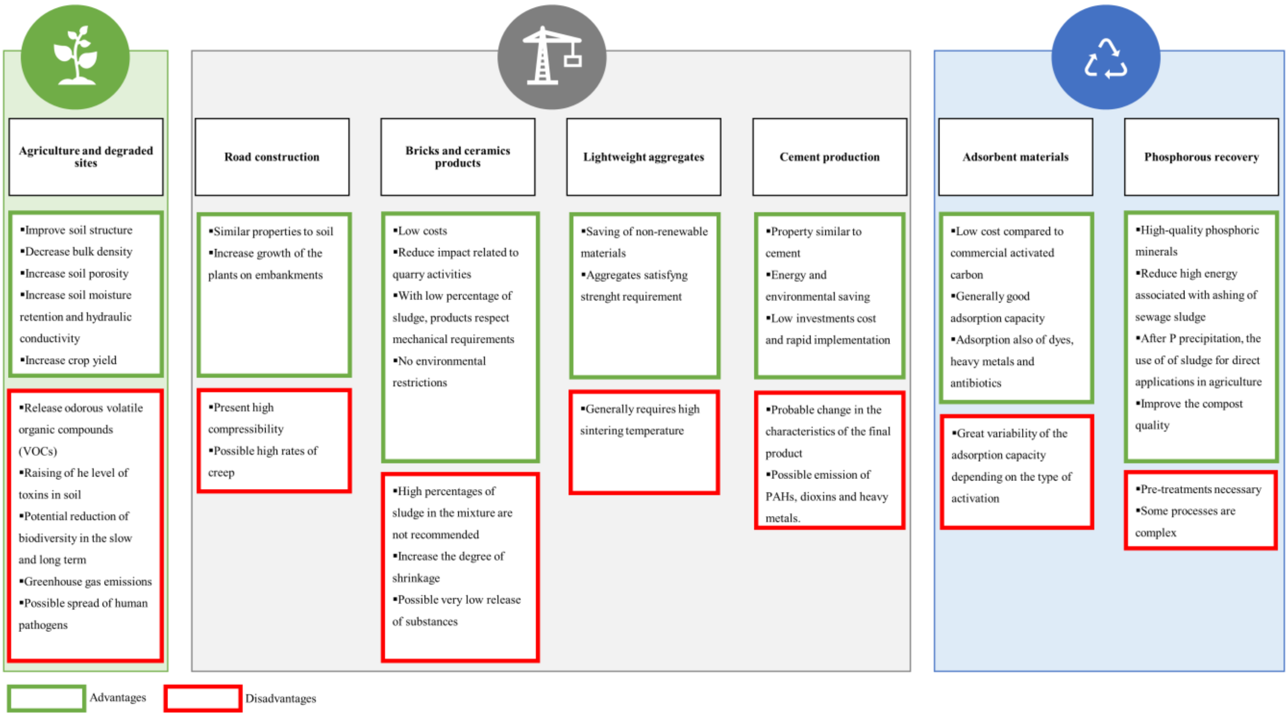
P-recovery from digested sludge is obtained also through wet-chemical process, applying extraction chemicals, pressure and temperature in relation to the starting material used. That approach provides adding a strong acid to decrease the pH in order to dissolve the initially bound of phosphorous. The amount of chemicals consumed depend on sludge characteristics (e.g., water content) and the P-recovery rate is associated to the operative parameters (Egle et al., 2015). The principal issue is concerned the metals dissolved during this wet-chemical extraction, that require an intensive use of chemicals for separate they before the metal ions and the phosphate product can be precipitated (Sartorius et al., 2011). Other questions from this approach are: (i) complexity of treatment due to sewage sludge composition (in particular from chemical precipitation with Fe or Al), (ii) possible production of waste (i.e. acidified sludge) that required further treatments and (iii) high chemicals consumption (wet chemical) and their costs (Egle et al., 2015).

In recent years, nutrients recovery from sewage sludge via crystallization was developed for the final production of magnesium ammonium phosphate (struvite) and calcium phosphate. In order to recovery P-nutrient via crystallization, a solubilization of P to release of phosphate to the supernatant is necessary (Tyagi and Lo, 2013). Up to 85% of dissolved P can be recovery from digested supernatant by crystallization or instant precipitation (Egle et al., 2016). (Fischer et al., 2011) described the phosphate extraction from digested sludge through microbial fuel cell (MFC), with energy production, by an E. coli cultivation: $FePO_4$ was reduced into MFC and the phosphate contained in the supernatant solution was precipitates as struvite through the addition of Mg^{2+} and NH_4^+.

The use of a P-recovery process also depends on the pollutant content in the sewage sludge (mainly heavy metals): wet-chemical leaching and wet oxidative approaches shows a depollution potential up to 98% of all heavy metals for sewage sludge (Egle et al., 2016).

Figure 1 shows the various possible options for recovery of biosolids. They are classified in three macro categories: agriculture and degraded sites (chapter 4), engineering applications (chapter 5) and other types of recovery (chapter 6). For each type of reuse the main advantages and disadvantages are reported.

Figure 1: *Biosolids recovery options and their respective advantages/disadvantages.* (Double-column figure) (COLOR)



7. Conclusions

This paper has sought to review positive and negative effects of recovery of sewage sludge on land, such as amendant/fertilizer, and for the production of cement, LWAs and bricks, where is used as a substitute of natural material.

As pointed out in this work, land application of biosolids improve soil properties, but requires further investigation, especially for effects connected with OCs. On the other side, as showed, the presence heavy metals and pathogens don't imply problems for human health because the first can be immobilized by some plants species protecting the food chain and human health, the second can be inactivated by different physicochemical and biological process. In opposition several authors highlighted than, in some cases, it is better to prevent the spreading on land for the consequence connected with human health.

Furthermore, studies have examined materials with sewage sludge, as a substitute of raw material, in engineering application, for instance in the road base layer, and sometimes as load bearing media. Results highlighted the good qualities of cement, LWAs and bricks, which did not show particular problems for human health. Obstacles to their use are due to opposition of population, which perceived the use of sewage sludge as dangerous for human health and for environment, and due to restring limit values imposed on waste, which are more restrictive than other materials.

As shown, many authors have demonstrated the feasibility, by a mechanical point of view, of the utilization of biosolids for brick construction, however some sectors of public opinion are against to put in practice that process. Although the pressure of population, the reuse of sewage sludge in the sector analysed is certainly interesting and would allow energy and environmental savings, emission reduction, immobilization of heavy metals, etc.

The conversion of biosolids into adsorbent material is also presented. In particular, the low cost of this production is highlighted compared to traditional adsorbent materials. Moreover, many studies have also shown the possibility of using biosolids as a source of P for a subsequent recovery. This is a very significant result that reduces the high energy associated with the incineration of sewage sludge, which is still a common practice in most European countries.

In conclusion, authors highlighted the importance of the continued vigilance and data collection because are indeed helpful in order to monitor and determine also the significance and implications of “emerging” organic contaminants for land application of biosolids. Further investigation would be conducted to understand effects of recovery of sewage sludge.

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none

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Figure

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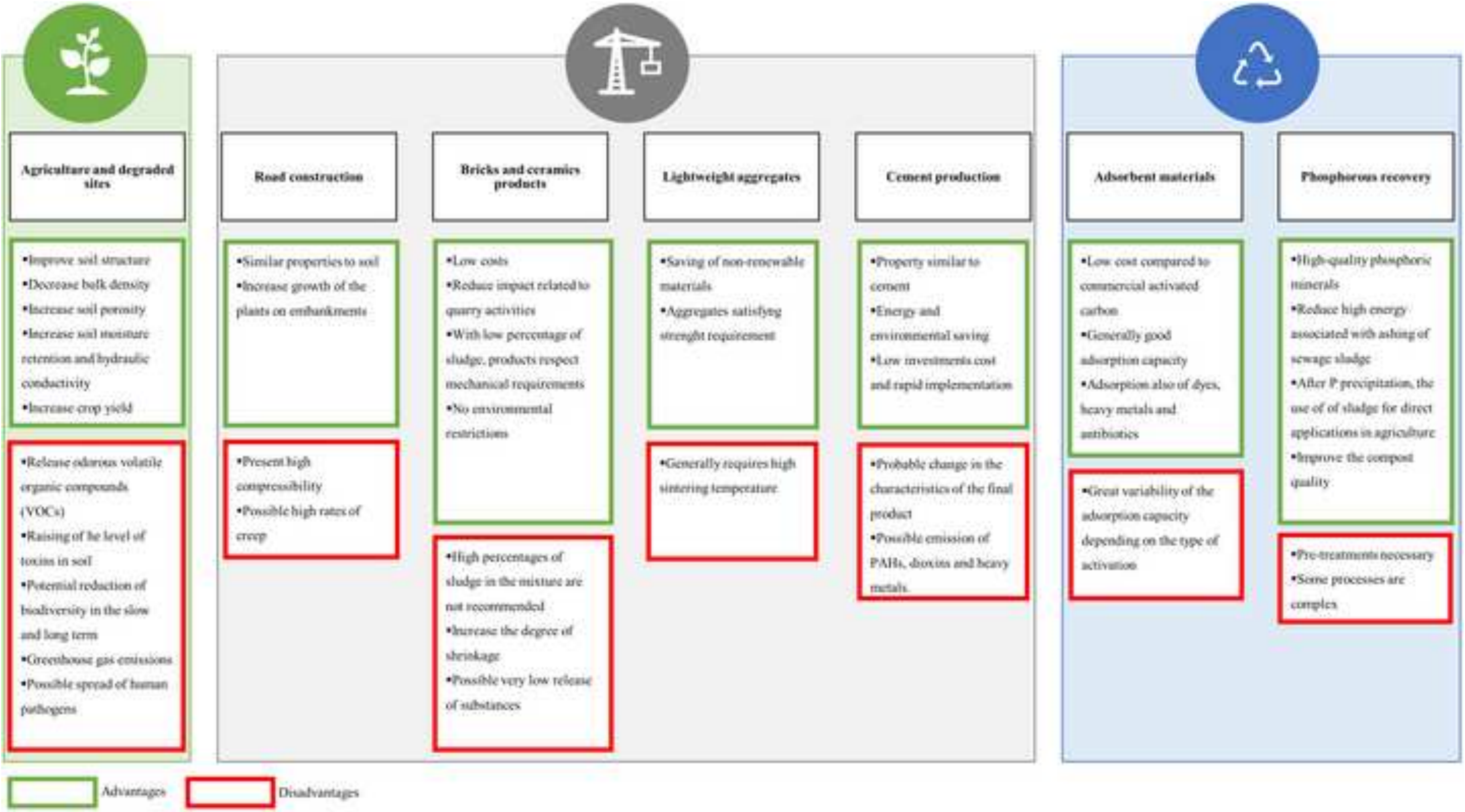

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