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Recycling sludge on cropland as fertilizer – advantages and risks

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Abstract

Background Digested sludge is a good source of plant nutrients. However, depending on the feedstock, it might contain heavy metals, metalloids, organic compounds, pathogens, and pharmaceuticals, which can cause adverse effects on crop growth and contaminate the groundwater, soil, and food chain.

Scope The aim of this review is to focus on the potential risks of inorganic and organic contaminants to plant growth, soil, groundwater, and consequently the food chain and environment related to the utilization of digested sludge as a fertilizer on cropland.

Conclusions Inorganic compounds, such as metals and metalloids, in sludge can occasionally cause reductions in soil microbial biomass. In general, the uptake of metals and organic contaminants does not appear to cause a significant hazard to the plants and the concentrations do not surpass the maximum values allowed in soil. Organic compounds, harmful for human health or the environment, are to a large extent decomposed or volatilized from the land treated with sludge, which decreases their leaching into the environment. Many of the organic compounds are lipophilic and can be bound to soil organic matter. In conclusion, the application of sludge on cropland might be a sustainable management practice; however, further investigations are needed to determine the accumulation and persistence of possible hazardous emerging chemicals and pathogens in the environment and formation of harmful intermediate reaction of inorganic and organic compound products.

Keywords Digestate, Food chain, Nutrient cycling, Pollutants, Sustainable agriculture

1. Introduction

The global population has increased rapidly from 5.3 billion in 1992 to 7.6 billion in 2018 and will reach 9.9 billion in 2050 (World Population Data Sheet, 2018). Such a rapid population growth will cause an increase in the consumption of water globally and consequently increase wastewater production along with digested sewage sludge, which represents about 0.3–0.5% of treated wastewater (Li et al., 2011). The disposal of digested sludge (herein sludge) in a safe way is a major environmental concern all over the world. The application of sludge on land after appropriate processing would support increased sustainability of agricultural production, as it recycles the nutrients back to the soil and makes them available to plants (Petersen et al., 2003; Antoline et al., 2005; Bozkurt et al., 2006; Seleiman et al. 2012, 2013a, 2013b; Bai et al., 2017; Seleiman et al., 2017).

Sludge is a solid or semi-solid by-product of domestic, industrial, and storm wastewaters treated through aerobic or anaerobic digestion processes in wastewater treatment plants (WWTPs) (Gardiner et al., 1995; Rogers, 1996; Andersen, 2001; Epstein, 2003; Harrison et al., 2006; Bianchini et al., 2016; Lovingood et al. 2018; Saleh Bairq et al., 2018). Sludge may also contain some inputs from farms, such as plant residues and manure (Fytili and Zabanioto, 2008; Seleiman et al., 2013a). The processing of wastewater includes primary (i.e., physical and/or chemical), secondary (i.e., biological), and finally tertiary (i.e., nutrient removal) treatments (Fytili and Zabanioto, 2008). Aerobic digestion is a process with a retention period of 7 days during which the sludge is subjected to at least 55 °C for an adequate period to ensure that the composting process is completed. In anaerobic digestion, primary digestion of 12 days at 35 °C or 24 days at 25 °C is followed by a retention period of at least 14 days (Tchobanoglous and Burton, 1991).

The use of sludge as a fertilizer can reduce the need for synthetic inorganic fertilizers (Seleiman et al., 2013a; Urbaniak et al., 2017) and can provide some micronutrients that are otherwise not added to the soil (Seleiman et al., 2012, 2013a, 2013b). Moreover, using sludge as a source of nutrients in agriculture can save non-renewable sources of energy for more sustainable production (Seleiman et al., 2013a; Urbaniak et al., 2017). Anaerobic digestion of wastewater sludge also produces methane, a valuable biofuel (Berktay and Nas, 2008; Gilbert et al., 2011). Sludge can be used as an alternative to synthetic fertilizers, for example for bioenergy crops (Seleiman et al., 2012, 2013a, 2013b; Urbaniak et al., 2017). This alternative use not only acts as an efficient method of sludge management, but is also in line with the implementation of the renewable energy directive 2009/28/EC, which requires 20% of total energy to be obtained from renewable sources in the European Union (EU) (Urbaniak et al., 2017).

In addition to beneficial plant nutrients, sludge may contain a variety of inorganic and organic substances, pharmaceuticals, and pathogens depending on the inputs of effluents in the wastewater plants and types of digestion used in the process (Carrington, 2001; Urbaniak et al., 2017; Hudcová et al., 2019). For instance, anaerobic digestion or even aerobic composting of sludge can be the main cause of organic contaminants, since the substances in the sludge may be partially biodegradable and new toxic intermediates can be formed (Schowanek et al., 2004). The main part of organic materials in sludge comes from human fecal material, but industrial catchment wastewaters can also be a source of organic material in sludge (Rogers, 1996; Andersen, 2001).

The benefits of adding any sludge as fertilizer to cropland must be compared with the risks of any contamination of the food chain by harmful substances that the sludge may contain and any leaching of the contaminants or plant nutrients to the environment. The awareness regarding inorganic and organic pollutants in food chain is increasing constantly, though knowledge gaps still remain. Therefore, it is of utmost importance to update the knowledge related to emerging and existing pollutants especially in sludge, since the interest to recycle it in croplands is constantly increasing. This article reviews the current understanding of the major risks to the food chain and the environment related to the use of sludge in agricultural soils.

2. Production and disposal of sludge

The alternatives for the disposal of sludge are agricultural use, composting, incineration, and landfill (Epstein, 2003; EUROSTAT, 2018; Hudcová et al., 2019). In the past, agricultural use (37% of the sludge produced in Europe) and landfill (40%) were considered the most economical options for sludge disposal (Tchobanoglous and Burton, 1991; Fytili and Zabaniotou, 2008), followed by incineration (11%) and some other uses (12%) such as forestry and land reclamation (Fytili and Zabaniotou, 2008). However, incineration (39%) and agricultural use (25%) are considered the main methods of sludge disposal in the EU nowadays, followed by composting (15%), other uses (13%), and finally landfill (8%) (EUROSTAT, 2018; Hudcová et al., 2019). In 2015, half or more of the sludge produced in Ireland (80%), Albania (77%), UK (74%), Spain (70%), Norway (62%), and Bulgaria (53%) was applied to cropland, whereas less than 5% of the sludge produced in Finland and Romania, and none in Greece, Slovenia, Slovakia, and Malta, was applied to cropland (Figures 1, 2) (EUROSTAT, 2018; Hudcová et al., 2019).

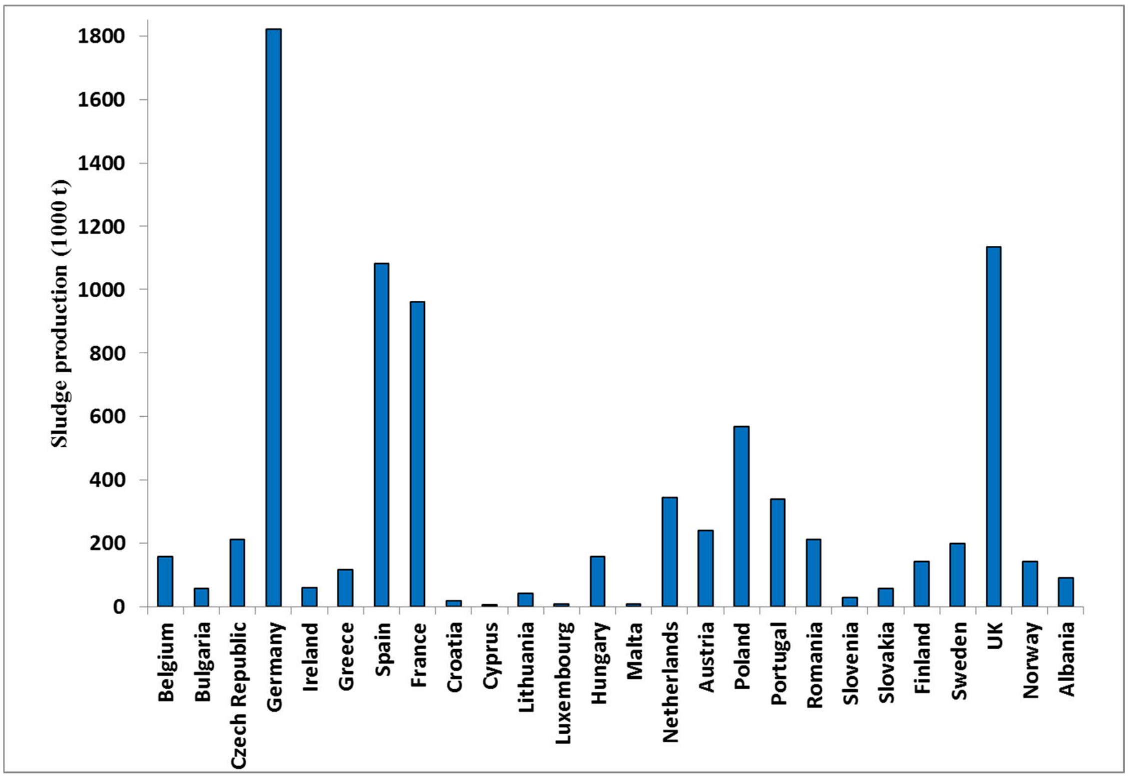


Figure 1

Annual production and agricultural use of sludge in European Union during 2012 – 2015 (EUROSTAT, 2018).

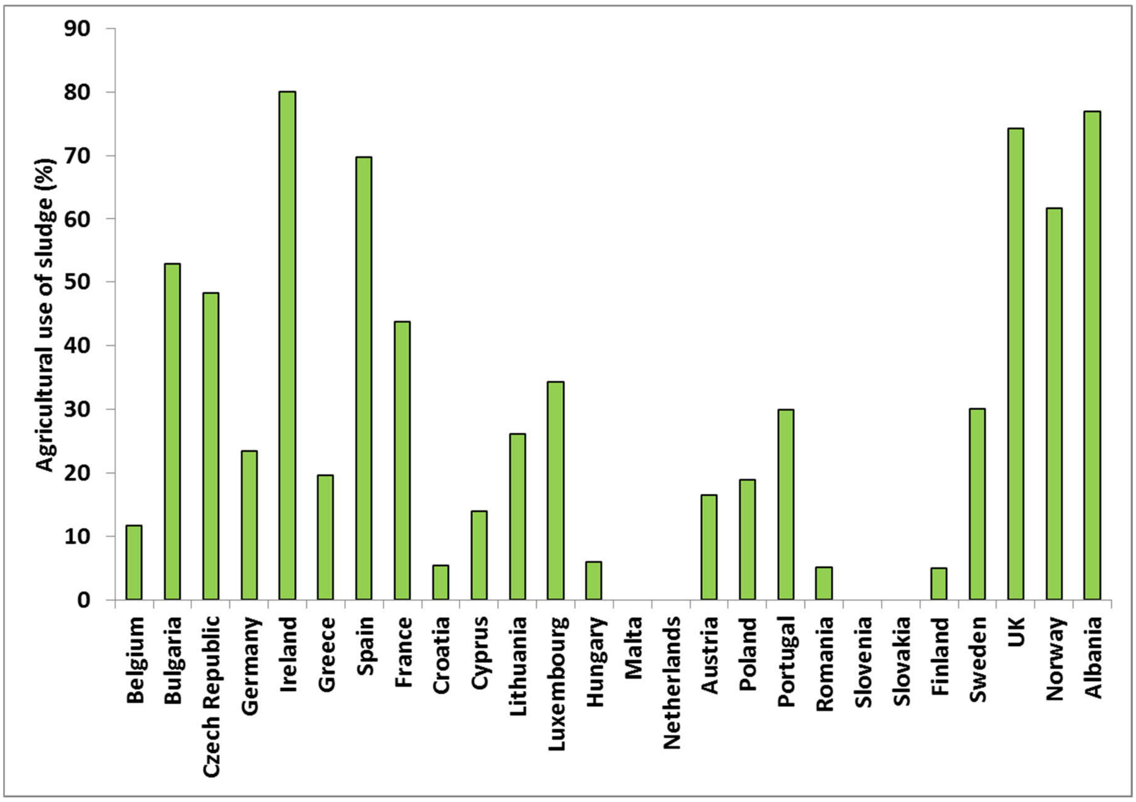


Figure 2

Agricultural use (%) of sludge in the European Union during 2012 – 2015 (EUROSTAT, 2018).

3. Composition of sludge

3.1. Inorganic compounds in sludge

Sludge contains nutrients, heavy metals, and metalloids (Seleiman et al., 2012, 2013a, 2013b; Bai et al., 2017; Fijalkowski et al., 2017; Rehman et al., 2018; Wang et al., 2018), some of which are essential macronutrients, such as nitrogen (N), phosphorus (P), potassium

(K), calcium (Ca), and manganese (Mn), and some are essential trace elements, such as boron (B), copper (Cu), iron (Fe), nickel (Ni), and zinc (Zn) (Table 1) (Epstein, 2003; Bozkurt et al., 2006; Kidd et al., 2007; Yan et al., 2009; Du et al., 2012; Gu et al., 2013; Seleiman et al., 2013a, 2017). However, at excessive concentration even the essential elements can be toxic similarly to non-beneficial heavy metals, such as cadmium (Cd), chromium (Cr), arsenic (As), and lead (Pb) (Pepper et al., 2006; Singh and Agrawal, 2008; Yan et al., 2009; Seleiman et al., 2013a).

The nutrient content of sludge varies markedly depending on the source of the wastewater, its treatment, as well as the sludge treatment and feedstock used (Table 1). Sludge can contain inorganic and organic N up to 51 g kg^{-1} dry matter (DM), and inorganic and organic P up to 28 g kg^{-1} DM (Yan et al., 2009; Seleiman et al., 2012, 2013a, 2013b; Gu et al., 2013). The bioavailability of P in sludge ranges from 40 to 80% (Andreoli et al., 2007), whereas the efficiency of synthetic P fertilizers is markedly lower, mostly due to the high soil capacity for fixing P (Mooso et al., 2013; Hudcová et al., 2019). In sludge, the content of K is much lower (less than 5 g kg^{-1} DM) than the content of N and P due to their high solubility (Antoline et al., 2005; Seleiman et al., 2013a). K compounds become mainly dissolved in the wastewater and do not settle with the sludge (Epstein, 2003; Yan et al., 2009). The content and bioavailability of N and P in sludge depends on the precipitation chemicals and the procedure used for cleaning wastewater (Table 1). As a comparison with other commonly utilized organic nutrient sources, animal manure (including decomposed straw, feces, and urine) contains approximately 1.2% N, 1.3% P, 1.5% K, 1.5% Mg, and 4.5% Ca, in addition to other nutrients (Xu et al., 2017), and meat-bone meal contains approximately 8.0% N, 4.4% P, 4.5% K, 1.5% Mg, and 10.0% Ca (Vamvuka et al., 2018).

Table 1

Examples of chemical properties of sludge of different origins. In Seleiman et al. (2013a), sludge was based either purely on sewage sludge or on sewage sludge digested with different feedstock materials like food leftovers and manure

	Pure sewage sludge	Digested sludge	Sewage sludge				
	Seleiman et al. (2013a)		Antoline et al. (2005)	Kidd et al. (2007)	Gu et al. (2013)	Grobelak et al. (2017)	Urbaniak et al. (2017)
pH	7.2	6.9	7.9	12.0	6.3	7.1	
C:N				9.5		47.4	
g kg^{-1} DM							
N	31.0	7.4	22.2	20.0	51.2	14.5	15.7
C				190.0		690.0	
P	26.0	9.9	16.6		5.51	3.5	13.3
K	2.1	1.1	4.7				
Ca	38.0			35.6			≥ 111.9
mg kg^{-1} DM							
Mn	220.0	56.9	226.0	350.0	130.0		
Cd	0.4	< 0.5	3.0	5.0	3.3	1.7	1.36
Cr	30.0	11.0	52.0	115.0	156.0		37.2
Cu	270.0	88.0	205.0	230.0	1120.0		55.8
Pb	20.0	4.9	80.5	69.0		149.0	≤ 26.1
Ni	20.0	5.5	25.0	35.0	52.8		≤ 10.4
Zn	470.0	130.0	731.0	500.0	30.0	288.9	344.8
As	5.0	1.3					

DM, dry matter

3.2. Organic compounds in sludge

Approximately 145 297 chemicals are preregistered in the European Chemicals Agency for industrial, agricultural, and household use (European Chemicals Agency, 2018). In the course of time, many of these substances will enter wastewater systems, where incompletely removed, hydrophobic compounds potentially remain in solid matter during water purification processes (Smith, 2009). In surveys from European and North American WWTPs, concentrations of organic compounds have varied from less than ng to g kg^{-1} DM of sludge depending on the source and processing techniques (Clarke and Smith, 2011). Regulations have decreased the concentration of most persistent, hazardous organic compounds in wastewaters, and they are no longer considered obstacles to the utilization of sludge in agriculture (Harrison et al., 2006).

Volatilized and biodegraded organic compounds can partially be lost through different treatment processes in wastewater plants via leaching, volatilization, and chemical and biological degradation in anaerobic or aerobic digestion (Smith and Riddell-Black, 2007). Nevertheless, anaerobic digestion and composting have not met in all cases the requirements and fixed limit concentrations set by the EU for the most commonly found organic pollutants in sludge, i.e., di-(2-ethylhexyl)phthalate (DEHP), nonylphenol ethoxylates (NPEs), linear alkylbenzene sulfonates (LASs), polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs), and polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs) (Stevens et al., 2003; Ahel et al., 1994). For example, surfactants LAS and NPE, and plasticizer DEHP, are aerobically biodegradable, but they are not degradable in mesophilic or thermophilic anaerobic digestion (Scott and Jones, 2000; IC Consultants, 2001).

Composting has been shown to reduce LAS compounds by 77–91% (Brunner et al., 1988; Pakou et al., 2009). Surfactants are amphiphilic and can be adsorbed into the organic matter of sludge and in addition metal ions may precipitate them into particulate matter, which is usually sedimented in WWTP settling tanks when they are not available for degrading microorganisms (Scott and Jones, 2000). PAHs, PCBs, and PCDD/Fs are hydrophobic and have low biodegradability. Thus, they tend to adsorb and accumulate in solid sludge (Fijalkowski et al., 2017; Hudcová et al., 2019). However, in aerobic soil environment surfactants may undergo further degradation (Scott and Jones, 2010).

Most pharmaceuticals found in influent are also present in sludge. Especially high concentrations of anti-inflammatory drug ibuprofen and some estrogens are adsorbed up to few mg kg^{-1} DM on sludge (Martín et al., 2012; Jelic et al., 2011). Degradation of pharmaceuticals in sewage treatment processes varies greatly: 41 pharmaceuticals from 21 different therapeutic groups accumulated in sludge up to concentrations of 100 ng g^{-1} DM (Jelic et al., 2011). Quite high concentrations of antimicrobial fluoroquinolones (e.g. 426 µG kg^{-1} Ciprofloxacin) are also known to adsorb into sludge, though those will be degraded in composting (Mitchell et al., 2015; Lillenberg et al., 2010).

Advanced oxidation technologies are proved to be promising methods to decompose organic pollutants from wastewater. Chemical, photochemical, sonochemical (ultrasound) and electroochemical processes either individually or in different combinations based in formation of very reactive, non-selective hydroxyl radicals initiate oxidative cascade leading to mineralization of organic pollutants to CO_2 and H_2O (Vanraes et al. 2016, Deng and Zhao 2015, Segneanu et al. 2013).

New biotechnological approaches have also been studied to degrade organic pollutants from the wastewaters and sludge. White-rot fungus (*Trametes versicolor* L.) is known to catalyze degradation of phenolic compounds (Addorisio et al., 2013; Catapane et al., 2013) as well as catabolize organic pollutants in non-sterile urban and hospital wastewater (Cruz-Morató et al., 2013) and sludge (Rodríquez-Rodríquez et al., 2011, 2012, 2014). However, in some cases new toxic intermediate compounds may form in catabolic reactions (Rodríquez-Rodríquez et al., 2012).

3.3. Pathogens in sludge

Sludge may also contain pathogens, such as bacteria, viruses, protozoa, and eggs of parasitic worms (Epstein, 2003; Yan et al., 2009; Urbaniak et al., 2017). Plant-based sludge could contain plant pathogens, such as viruses, bacteria, fungi, and parasites, along with undesired weed seeds (Carrington, 2001). Sludge of animal origin, such as from livestock production, slaughterhouses, or meat processing industries, could contain antibiotic resistant bacteria and viruses (Franke-Whittle and Insam, 2013). Slaughter wastes may also contain the prions of bovine spongiform encephalopathy (Carrington, 2001; Franke-Whittle and Insam, 2013). Typical concentrations of colony forming units of most common bacteria in untreated sludge are 10^2 to 10^3 g^{-1} DM of *Salmonella spp*. to 10^6 g^{-1} of *Escherichia coli*, 10^2 to 10^4 g^{-1} of enteroviruses, 10^2 to 10^3 g^{-1} of protozoa, and 5 to 10^3 g^{-1} of helminth eggs (Carrington, 2001). To produce sludge for safe use as fertilizer in agriculture, the treatment must reduce the number of *Salmonella spp*. and enteroviruses by at least four orders of magnitude as well as destroy the viability of Ascaris and helminths’ ova. In addition, the level of *Escherichia coli* should not exceed 1000 colony forming units g^{-1} DM sludge, and the level of

Clostridium perfringens spores should not exceed 3000 colony forming units g^{-1} DM (Strauch, 1998).

Pathogen level and diversity in wastewater vary temporally and geographically. Therefore, their continuous detailed monitoring is impossible. Continuous detailed monitoring can be replaced by the use of non-pathogenic surrogate organisms, which are sensitive indicators of virulent human pathogens (Carrington, 2001). Organisms used in validation processes are widespread in sludge and easy to maintain for validation tests *Escherichia coli* have proven to be a suitable indicator by having similar characteristics to vegetative bacteria, for example *Salmonella spp.*, *Shigella spp.*, *Listeria spp.*, and most viruses (Pike et al., 1988; Strauch, 1998).

Recycling sludge safely in agriculture requires environmentally and ecologically sustainable technologies in efficiently remove hazardous organic pollutants, pharmaceuticals, pathogens, and parasites from sludge. Common methods to stabilize and sanitize sludge in WWTPs are biological anaerobic and aerobic digestion, heat treatment and alkaline stabilization (Arthurson, et al. 2008; Goldfarb et al. 1999). Lime can be used to sanitize and disinfect sludge, and remove different pathogens, particularly bacteria (Urbaniak et al., 2017). Pathogenic microorganisms, which are found in sludge containing fecal and vegetable material, have been listed in the European Commission’s Evaluation of Sludge Treatments for Pathogen Reduction – Final Report No: CO 5026/1 (Carrington, 2001).

4. Sludge in agriculture and regulations regarding its use

4.1. Sludge on cropland

Sludge added to cropland has many potentially beneficial impacts due to the improvement of biological, chemical, and physical properties of soils, which may improve plant growth and productivity (Beck et al., 1996; Seleiman et al., 2012, 2013b; Bai et al., 2017; Urbaniak et al., 2017; Rehman et al., 2018; Hudcová et al., 2019). The high organic matter content may improve the functioning of sludge as a soil conditioner by increasing soil water-holding capacity and water infiltration, stabilizing soil temperature fluctuation, serving as a storage of nutrients, and enhancing soil microbial activity (Jarausch-Wehrheim et al., 1999; Epstein, 2003; Samaras et al., 2008; Yan et al., 2009; Seleiman et al., 2013b).

On the other hand, many soil factors, such as pH, organic matter, aeration, cation exchange capacity, water content, temperature, and elemental interactions, can affect the uptake of elements from the soil (Epstein, 2003). Soil pH is considered the main factor affecting the solubility of trace elements. For instance, the solubility of all essential trace elements except molybdenum (Mo) and selenium (Se) is increased at low pH; hence their potential uptake by plants increases. Depending on the environmental conditions, soil organic matter can reduce or increase the plant availability of cationic trace elements, such as Cd, Cu, Ni, and Zn, through chelate formation. High cation exchange capacity reduces the mobility of trace elements, such as Cd, Cu, Ni, and Zn. Due to the high cation exchange capacity of clay minerals, the binding of trace elements in clay soil is higher than in sandy soil (Epstein, 2003).

Similarly, soil organic matter has, on a mass basis, higher cation exchange capacity than the mineral fraction of soil. The interactions between macronutrients and trace elements can reduce the bioavailability of micronutrients in soil. For example, the interaction between the phosphate and trace elements can form soluble or insoluble compounds depending on soil pH (Epstein and Chaney, 1978). The less than optimal ratios of plant nutrient content in sludge are generally complemented by adding commercial fertilizers with sludge to cropland to balance the nutrition needed for each species (Yan et al., 2009) (Table 2).

Table 2

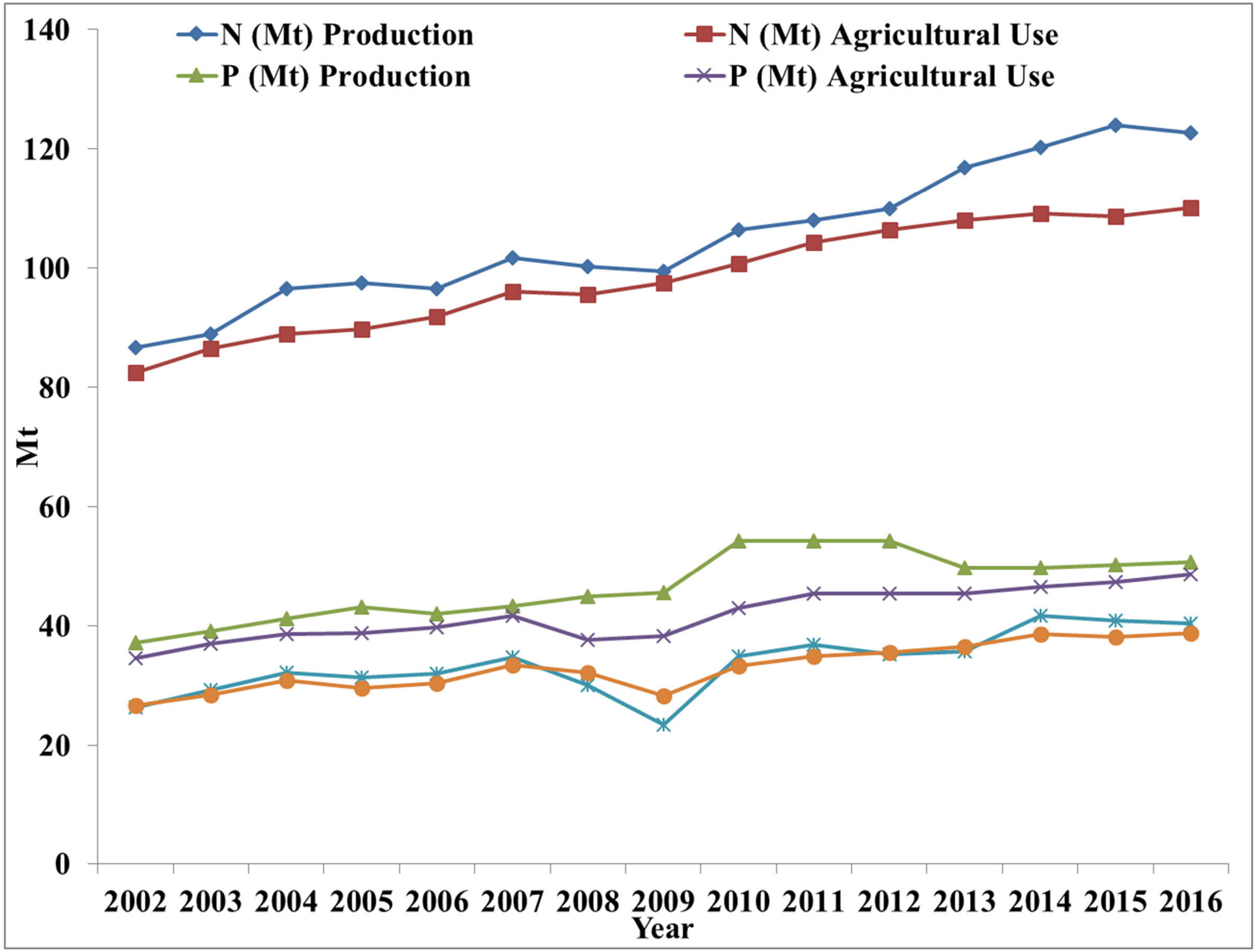
Doses of sludge applications as fertilizer for different plant species grown on cropland

Sludge type	Dose (t ha^{-1})	Plant species	Soil type	Authors
Sewage sludge	3.6, 7.2, 10.8, 14.4, 18.0	*Salix discolor* Mtihl. *Salix viminalis* L.	Sandy soil	Labrecque et al. (1995)
Sewage sludge compost	5.9, 11.7, 29.2, 58.5	*Brassica chinensis* L.	Loamy soil	Wong et al. (1996)
Municipal sewage sludge	0, 5, 7.5, 10, 15, 20	*Triticum aestivum* L.	Coarse loamy sand	Merrington et al. (1997)
Digested sewage sludge	10, 20, 30, 40	*Phaseolus vulgaris* L.	–	Wen et al. (1997)
Sewage sludge	10, 100	*Zea mays* L.	Acid sandy soil	Jarausch-Wehrheim et al. (1999)
Dried sewage sludge	10, 20, 30, 40, 50	*Zea mays* L.	–	Qasim et al. (2001)
Sewage sludge	60	*Diplotaxis erucoides* L.	Clayey and silty soil	Korboulewsky et al. (2002)
Anaerobically digested and activated sewage sludge	3.4	*Avena sativa* L.	Sandy loam soil	Petersen et al. (2003)
Sewage sludge	7, 14, 21	*Sorghum vulgare* L.	Sandy clay soils	Akdeniz et al. (2006)
Activated sludge and facultative stabilization ponds	100	*Sorghum vulgare* L.	Clay loam soil, sandy loam, loam	Mendoza et al. (2006)
Sewage sludge	10, 30, 50	*Gossypium hirsutum* L.	Typic Xerochrept (clay loam)	Samaras et al. (2008)
Sewage sludge	0, 15, 30, 60, 120	*Zoysia japonica*, *Poa annua*	Meadow brown soils	Wang et al. (2008)
Sewage sludge	9.8, 19.6	*Zea mays* L.	Chernozem	Černý et al. (2012)
Digested sludge	0.9 up to 16.2	*Zea mays* L. *Cannabis sativa* L. *Brassica napus* L.	Vertic Stagnosols	Seleiman et al. (2013a)
Sewage sludge	3 and 9	*Salix viminalis L.*	Sandy clay soils	Urbaniak et al. (2017)
Sewage sludge	1%	*Triticum aestivum L.*	Clay loam soils	Rehman et al. (2018)
Sewage sludge	0, 25, 50, 125, and 250	*Sorghum bicolor L.*	Halaquepts of inceptisols	Zuo et al. (2019)

According to Antoline et al. (2005) and Mendoza et al. (2006), sludge application increased the ammonium-N content, cation exchange capacity, and organic carbon content in soil. Unfortunately, sludge application also reduced soil pH and increased diethylenetriaminepentaacetic acid-extractable heavy metals, i.e., Cd, Cu, Zn, Pb, and Mn (Antoline et al., 2005; Mendoza et al., 2006). Moreover, the narrow C:N ratio of sludge could also accelerate organic matter decomposition, which could result not only in a loss of organic matter but also a gradual release of heavy metals and metalloids into the soil solution, increasing their availability to plants (Wołejko et al., 2014).

When applied as fertilizer, sludge increased the yield of alfalfa (*Medicago sativa* L.), wheat (*Triticum aestivum* L.), faba bean (*Vicia faba* L.), zoysia grass (*Zoysia japonica* L.), and biomass of maize (*Zea mays* L.), hemp (*Cannabis sativa* L.), oilseed rape (*Brassica napus* L. ssp. *oleifera* [Moench.] Metzg.), willow (*Salix viminalis* L.), and sweet sorghum (*Sorghum bicolor* L.) (Elsokkary and El-Keiy, 1988; Wang et al., 2008; Seleiman et al., 2012, 2013a; Urbaniak et al., 2017; Rehmana et al., 2018; Zuo et al., 2019). However, it also increased the heavy metal and metalloid content in the leaves and grains of barley (*Hordeum vulgare* L.) (Antoline et al., 2005) and sorghum leaves (*Sorghum bicolor*. L.) (Mendoza et al., 2006), as well as the biomass of maize, hemp, oilseed rape (Seleiman et al., 2012, 2013a, 2013b), and sweet sorghum (Zuo et al., 2019), without any visual symptoms of heavy metal toxicity.

Apart from improving soil structure and nutrient recycling, agricultural use of sludge also has the potential to save energy compared with synthetic fertilizers. Almost 40% of the commercial energy used in agriculture is consumed for manufacturing synthetic N fertilizers due to the large energy input required for reducing N_2 to ammonia in the Haber–Bosch process (Mudahar and Hignett, 1985; Helsel, 1992). The energy needed to produce synthetic fertilizers including the packaging, transportation, and application to the cropland is about 78 MJ kg^{-1} N, 17 MJ kg^{-1} P, and 14 MJ kg^{-1} K (Helsel, 1992; Elsayed and Mortimer, 2001; Mikkola and Ahokas, 2009). Production of N fertilizers increased from 11.6 to 122.7 Mt and synthetic P fertilizers from 10.0 to 50.8 Mt during the period 1961–2016 (IFASTAT, 2016; FAOSTAT, 2012, 2019). During the period 2002–2016, the agricultural use of N fertilizers increased from 82.5 to 110.2 Mt and the use of P fertilizers from 34.6 to 48.6 Mt (Figure 3) (FAOSTAT, 2019).


N (Mt) Production
N (Mt) Agricultural Use
P (Mt) Production
P (Mt) Agricultural Use
Mt
0
20
40
60
80
100
120
140
2002
2003
2004
2005
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2007
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Figure 3

World production and agricultural use of nitrogen (N), phosphorus (P), and potassium (K) fertilizers during 2002–2016 (FAOSTAT, 2019).

4.2. Main regulations related to use of sludge on cropland

Sludge can contain, depending on the source and processing technique in WWTPs, a quantitatively and qualitatively variable range of harmful compounds, pathogens, and pharmaceuticals (Carrington, 2001; Clark and Smith 2011; Seleiman et al., 2013a; Al-Gheethi et al., 2018; Hudcová et al., 2019). Therefore, its use on land is usually regulated.

The amount of heavy metals in sludge as well as in soil to which sludge is applied is strictly regulated in the EU (Table 3). Council Directive 86/278/EEC of 12 June 1986 aimed to protect the environment and prevent the harmful effects of adding sludge to cropland to plants, animals, and humans (European Commission, 1986). The directive restricts: 1) contamination of the soil by heavy metals and metalloids by limiting the contents of heavy metals and metalloids in the soil to which sludge is applied, 2) the content of heavy metals and metalloids in sludge, as well as 3) the amounts of heavy metals that can be annually added to the soil (Table 3) and the maximum number of pathogens in sludge (European Commission, 1986). Some EU countries have more restrictive requirements compared with Directive 86/278/EEC and have approved the limits for heavy metal concentrations, synthetic organic compounds, and microbial contamination (Hudcová et al., 2019). For example, in the Netherlands the limits of Cd, Cu, Ni, Pb and Zn concentrations in sludge should not exceed 1.2, 75, 30, 100, and 300 mg kg^{-1}, respectively (EUROSTAT, 2018; Hudcová et al., 2019).

In the EU, specific requirements for organic compounds in sludge are not included in Directive 86/278/EEC, but to reduce potential health risks several national regulations include limitations concerning the allowable amounts of organic compounds in the sludge applied to cropland. For example, the limits of PAHs and PCBs were 6.5 and 1.0 mg kg^{-1} DM sludge in Bulgaria, respectively, while the limits of DEHP, LASs, NPEs, and PAHs were 50, 300, 10, and 3 mg kg^{-1} DM sludge in Denmark, respectively. In addition, the limits of adsorbed organic halogen compounds (AOX) and PCBs were 500 and 0.2 mg kg^{-1} DM sludge in Germany, respectively (European Commission, 2000; Hudcová et al., 2019).

Table 3

European Union limits for heavy metals in sludge and soil to which sludge is applied (European Commission, 1986, 2001, 2002)

	Cd	Ni	Cu	Cr	Pb	Zn
Soil treated with sludge (mg kg^{-1} DM)	3	75	140	150	300	300
Previous regulations(before 2015)						
Heavy metals in sludge (mg kg^{-1} DM)	40	400	1750	1000	1200	4000
Maximum load of heavy metals to agricultural soil (g ha^{-1} year^{-1})	150	3000	12000	3000	1500	30000
Current regulations from 2015						
Heavy metals in sludge (mg kg^{-1} DM)	5	200	800	800	1500	2000
Maximum load of heavy metals to agricultural soil (g ha^{-1} year^{-1})	15	600	2400	2400	1500	6000
Proposed regulations 2025						
Heavy metals in sludge (mg kg^{-1} DM)	2	100	600	600	200	1500
Maximum load of heavy metals to agricultural soil (g ha^{-1} year^{-1})	6	300	1800	1800	600	4500

DM, dry matter

In the USA, the Environmental Protection Agency’s regulation 40 CFR Part 503 Standards for the Use or Disposal of Sewage Sludge (U.S. Federal Register, 1993) defines two sludge classes: Class A and Class B. Class A sludge has undergone composting, heat drying, and high-temperature aerobic digestion, which reduces pathogenic bacteria, enteric viruses, and viable helminths’ ova to below detectable levels. Class A sludge can be used as a soil amendment without imposing site and harvesting restrictions. Class B sludge may still contain some pathogens, and thus its application is restricted for the harvestable crops, animal grazing, and public access for a period of time after application (U.S. Federal Register, 1993).

Sludge application is also limited in many European countries either as a maximum amount ha^{-1} year^{-1} or by P-based agronomic rates. In the USA, a N-based application rate is used and no specific limitations for organic contaminants exist (Harrison et al., 2006).

5. Risk of contamination of food chain and the environment

Potential adverse effects of toxic inorganic and organic compounds on the environment and living organisms through the food chain follow the application of sludge to cropland (Epstein, 2003; Schowanek et al., 2004; Bai et al., 2017). Sludge can pose a risk to the surface water and groundwater (Rlöpffer, 1996; Grobelak et al., 2017), the environment, and thus the food chain (Duarte-Davidson and Jones, 1996). Pathogens in sludge can pose risks to human health if transferred to food crops grown on sludge-treated soils (Yan et al., 2009; Urbaniak et al., 2017; Hudcová et al., 2019).

Even though sludge is a valuable source of nutrients, for example excessive P accumulation in soil as a source of pollution for surface water and groundwater is of concern (Grobelak et al., 2017). This can be caused by the non-optimal ratios of plant nutrients in sludge. Sludge application at recommended rates based on the content of available soil P might result in an accumulation of P in soil, which can increase the risk of eutrophication and an adverse impact on water bodies through surface runoff, subsurface drainage water, and eroded soil. To avoid leaching of P into the environment, wastewater treatment facilities have been required to meet stricter effluent limits for P (Łuczkiewicz, 2006). Sludge produced in WWTPs, where Fe, aluminum (Al), or Ca are used during pretreatment to reduce the soluble P (to meet effluent limits), also has lower P available to plants (i.e., less than 25% of that in triple superphosphate). Phosphate is strongly adsorbed to the surfaces of Fe and Al hydrous oxides and calcium carbonate (Bastin et al., 1999). In addition, heat-dried sludge has low P availability.

Heavy metals and metalloids are non-biodegradable and can be taken up by plant roots and stored in different plant tissues (Wagner, 1993; McLaughlin et al., 1999). The possibility of their accumulation in human tissues and biomagnification via the food chain can cause risks to human health and the environment (Krogmann et al., 1999). The mobility of heavy metals, their bioavailability, and their link to ecotoxicity are based on their specific chemical forms and the mechanisms of binding (Fuentes et al., 2004).

Fuentes et al. (2004) investigated four different products of sludge deriving from different wastewater plant treatments (i.e., aerobic, anaerobic, unstabilized, and sludge from a waste stabilization pond) to compare the influence of the stabilization method on the distribution of heavy metals. Based on that, anaerobic sludge should not be applied to cropland due to its high Cr content. Sludge from a waste stabilization pond, which had a higher level of mineralization and stabilization than the other products of sludge, had a lower heavy metal availability index. This could be due to a correlation between the mobile oxidizable and residual fractions. In contrast, the unstabilized sludge had the highest heavy metal accumulation in the most easily assimilable fractions (bioavailable, exchangeable, and reducible) (Fuentes et al., 2004).

A high content of persistent and bioaccumulative organic contaminants in sludge is a concern when sludge is used on cropland and in landfills, which may result in the spread of organic pollutants into the environment and food chain. Many organic pollutants include a number of congeners with different chemical quality and toxicities (Hudcová et al., 2019). The concentrations of many of those “traditional” organic contaminants in sludge is regulated by source control, but there exist still vast number of emerging potential compounds whose toxicity and behavior in soil and water bodies have not been analyzed yet (Eriksson et al. 2008).

Temperature and time affect the degradation rate of LAS and NPE compounds in soil, which must be considered when timing sludge storage and application to soil. For example, the half-life of LAS at 22 °C was 7 days in comparison with 14 days at 13 °C (Gonzaléz et al., 2010), and it took 56 days for NPE and 23 days for LAS to lose toxicity in the soil (Garrido-Perez et al., 2008). Thus, LAS and NPE compounds can be loaded into soil via sludge and transferred into surface water and groundwater. In the environment, NPEs degrade to nonylphenol (NP) which is more persistent and toxic especially in aquatic environment (U.S. Environmental Protection Agency, 2010).

These organic xenobiotics can also inhibit processes like degradation of organic matter and mineralization (Kümmerer, 2009; Eriksson et al., 2008). If it accumulates in the food chain, NPE as a pseudo-estrogen will disrupt the animal and human endocrine system.

Clarke and Smith (2011) ranked selected emerging organic compounds using an assessment matrix approach with five properties: 1) environmental persistence (> 6 months), 2) human toxicity, 3) bioaccumulation, 4) ecotoxicity, and 5) number and quality of international studies of the contaminant. On the basis of the results, the organic compounds were organized in decreasing priority as follows: perfluorinated chemicals (PFCs) > polychlorinated alkanes (PCAs) and polychlorinated naphthalenes (PCNs) > organotins, polybrominated diphenyl ethers, triclosan and triclocarban > benzothiazoles > antibiotics and pharmaceuticals > synthetic musks > bisphenol A and phthalate acid esters and quaternary ammonium compounds > steroids, and polydimethylsiloxanes.

In hazard identification analysis of including 192 organic compounds commonly found in sludge, 51,6% were classified as hazardous, 12,5% as non-hazardous and 32,8% of the compounds could not be classified due to the lack of basic data, such as biodegradation, short-term and chronic toxicity for the aquatic and soil organisms (Eriksson et al., 2005).

Short-chain perfluorochemicals (PFCs), perfluorooctanoic acid (PFOA), perfluorooctane sulfonates (PFOS), and polychlorinated alkanes (PCAs) have caused concern. All these substances have been found in human blood (Olsen et al., 2003), human milk (Thomas et al., 2006), and the environment (Campbell and McConnell, 1980; Giesy and Kannan, 2001). PCAs are persistent and bioaccumulative and their mean concentration of sludge collected from WWTPs in UK was much higher, 1800 mg kg^{-1} DM in comparison to other persistent organic pollutants (Stevens et al 2003). Short-chain PCAs and PFOAs are identified as priority hazardous substances by European Water Framework directive and placed restrictions on marketing and use (REACH Annex XVII).

Uptake of organic compounds by plants depends on the lipophilicity and water solubility of the substance, the ambient temperature, as well as the concentration of organic compounds in the soil and plants (Simonich and Hites, 1995). Organic compounds are taken up from a soil solution by roots and translocated into the leaves. Some of these compounds can be metabolized during the uptake and some intermediates can be translocated from the roots to other plant parts, while others can remain in the plant roots. Volatile organic contaminants can enter foliage through the wax-containing cuticle, which can bind and store lipophilic organic compounds, as well as through the stomata (Duarte-Davidson and Jones, 1996). Maize uptake of PFCs from nutrient medium was dependent on chain length of substances. For instance, short-chain substances were taken up more actively than long-chain substances, and translocated to shoot. Whereas higher concentration of long-chain substances, for example PFOA and PFOS, accumulated in roots (Krippner et al. 2014). Wheat and earthworms (*Eisenia fetida* L.) take up PFOS precursor perfluorooctane sulfonamide, which is transformed to PFOS in both organisms and other derivatives in wheat (Zhao et al. 2018). Organic compounds in soil treated with sludge can translocate and accumulate in grazing livestock, depending on some factors such as livestock species, type of sludge applied to the soil, and growing season. For example, PAHs could adhere to plant root surfaces and the lighter molecular weight compounds might volatilize from polluted soil into the foliage. Grazing livestock can ingest the organic compounds in three different ways: (a) grazing, (b) adhering to soil and/or sludge with harvested forage, and (c) ingestion of the soil–sludge mixture. For instance, up to 2% of a sheep’s diet is soil (Duarte-Davidson and Jones, 1996). Nonpolar and persistent organic compounds can be stored and accumulated in fat tissues and milk if not metabolized (Wild and Jones, 1992). Other organic compounds such as PCDD/Fs, PCBs, and hexachlorobenzene are not metabolized (Geyer et al., 1987). In fact, the potential accumulation of PCDD/Fs in livestock is a critical issue, since about two thirds of humans get PCDD/Fs from animal fat – mostly dairy products and beef (Fürst et al., 1990; Beck et al., 1992).

The livestock industry, manure, as well as industrial and hospital wastewater can contain zoonotic and non-zoonotic pathogens and antimicrobial-resistant bacteria. Mesophilic treatment or even short thermophilic treatment is not always enough to eliminate pathogens from sludge. The samples from raw sludge (67%) and digested sludge (55%) were positive for different *Salmonella spp*. serotypes as well as *Campylobacter* (2%) and *Listeria* (4%) after a 9-day thermophilic and 28-day mesophilic treatment (Sahlström et al., 2004). In general, an over 10-day thermophilic treatment is required for appropriate sanitation of *Escherichia coli* and *Salmonella* spp. in sludge. However, spores of *C. perfringens* were not destroyed even in a 15-day thermophilic treatment (Coelho et al., 2011; Lloret et al., 2013). The safe recycling of sludge in agriculture requires that contaminants and pollutants accumulating from wastewaters into sludge are identified and environmental risk assessments made to determine the limits for these contaminants in sludge intended for agricultural purposes (Andersen, 2001).

6. Conclusions

Even though sludge seems to be a good source of nutrients and improves the level of nutrient recycling, the application of sludge to cropland should not adversely affect groundwater or the food chain. Before sludge recycling in agriculture can be widely accepted, more information about possible interactions between inorganic and organic compounds and their intermediate reaction products, their accumulation and persistence in the environment, is required. To date, few investigations have been done on the leaching of organic pollutants when sludge is applied to soil. Further investigations are needed on the residual organic chemicals remaining in the soil or taken up by plants when sludge is applied to cropland, as such work is important for assessing the risks posed to the environment and food chain.

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