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Elsevier B.V.

2020-04

Seleiman, M, Santanen, A & Mäkelä, P 2020, 'Recycling sludge on cropland as fertilizer – advantages and risks', *Resources, Conservation and Recycling*, vol. 155, 104647. <https://doi.org/10.1016/j.resconrec.2019.104647>

<http://hdl.handle.net/10138/337795>

10.1016/j.resconrec.2019.104647

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1 **Recycling sludge on cropland as fertilizer – advantages and risks**

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10

11 **Abstract**

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

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

19 *Conclusions* Inorganic compounds, such as metals and metalloids, in sludge can
20 occasionally cause reductions in soil microbial biomass. In general, the uptake of metals and
21 organic contaminants does not appear to cause a significant hazard to the plants and the
22 concentrations do not surpass the maximum values allowed in soil. Organic compounds,

23 harmful for human health or the environment, are to a large extent decomposed or volatilized
24 from the land treated with sludge, which decreases their leaching into the environment. Many
25 of the organic compounds are lipophilic and can be bound to soil organic matter. In
26 conclusion, the application of sludge on cropland might be a sustainable management
27 practice; however, further investigations are needed to determine the accumulation and
28 persistence of possible hazardous emerging chemicals and pathogens in the environment and
29 formation of harmful intermediate reaction of inorganic and organic compound products.

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

31 **1. Introduction**

32 The global population has increased rapidly from 5.3 billion in 1992 to 7.6 billion in 2018
33 and will reach 9.9 billion in 2050 (World Population Data Sheet, 2018). Such a rapid
34 population growth will cause an increase in the consumption of water globally and
35 consequently increase wastewater production along with digested sewage sludge, which
36 represents about 0.3–0.5% of treated wastewater (Li et al., 2011). The disposal of digested
37 sludge (herein sludge) in a safe way is a major environmental concern all over the world.
38 The application of sludge on land after appropriate processing would support increased
39 sustainability of agricultural production, as it recycles the nutrients back to the soil and
40 makes them available to plants (Petersen et al., 2003; Antoline et al., 2005; Bozkurt et al.,
41 2006; Seleiman et al. 2012, 2013a, 2013b; Bai et al., 2017; Seleiman et al., 2017).

42 Sludge is a solid or semi-solid by-product of domestic, industrial, and storm wastewaters
43 treated through aerobic or anaerobic digestion processes in wastewater treatment plants
44 (WWTPs) (Gardiner et al., 1995; Rogers, 1996; Andersen, 2001; Epstein, 2003; Harrison et

45 al., 2006; Bianchini et al., 2016; Lovingood et al. 2018; Saleh Bairq et al., 2018). Sludge
46 may also contain some inputs from farms, such as plant residues and manure (Fytili and
47 Zabanioto, 2008; Seleiman et al., 2013a). The processing of wastewater includes primary
48 (i.e., physical and/or chemical), secondary (i.e., biological), and finally tertiary (i.e., nutrient
49 removal) treatments (Fytili and Zabanioto, 2008). Aerobic digestion is a process with a
50 retention period of 7 days during which the sludge is subjected to at least 55 °C for an
51 adequate period to ensure that the composting process is completed. In anaerobic digestion,
52 primary digestion of 12 days at 35 °C or 24 days at 25 °C is followed by a retention period
53 of at least 14 days (Tchobanoglous and Burton, 1991).

54 The use of sludge as a fertilizer can reduce the need for synthetic inorganic fertilizers
55 (Seleiman et al., 2013a; Urbaniak et al., 2017) and can provide some micronutrients that are
56 otherwise not added to the soil (Seleiman et al., 2012, 2013a, 2013b). Moreover, using
57 sludge as a source of nutrients in agriculture can save non-renewable sources of energy for
58 more sustainable production (Seleiman et al., 2013a; Urbaniak et al., 2017). Anaerobic
59 digestion of wastewater sludge also produces methane, a valuable biofuel (Berkay and Nas,
60 2008; Gilbert et al., 2011). Sludge can be used as an alternative to synthetic fertilizers, for
61 example for bioenergy crops (Seleiman et al., 2012, 2013a, 2013b; Urbaniak et al., 2017).
62 This alternative use not only acts as an efficient method of sludge management, but is also
63 in line with the implementation of the renewable energy directive 2009/28/EC, which
64 requires 20% of total energy to be obtained from renewable sources in the European Union
65 (EU) (Urbaniak et al., 2017).

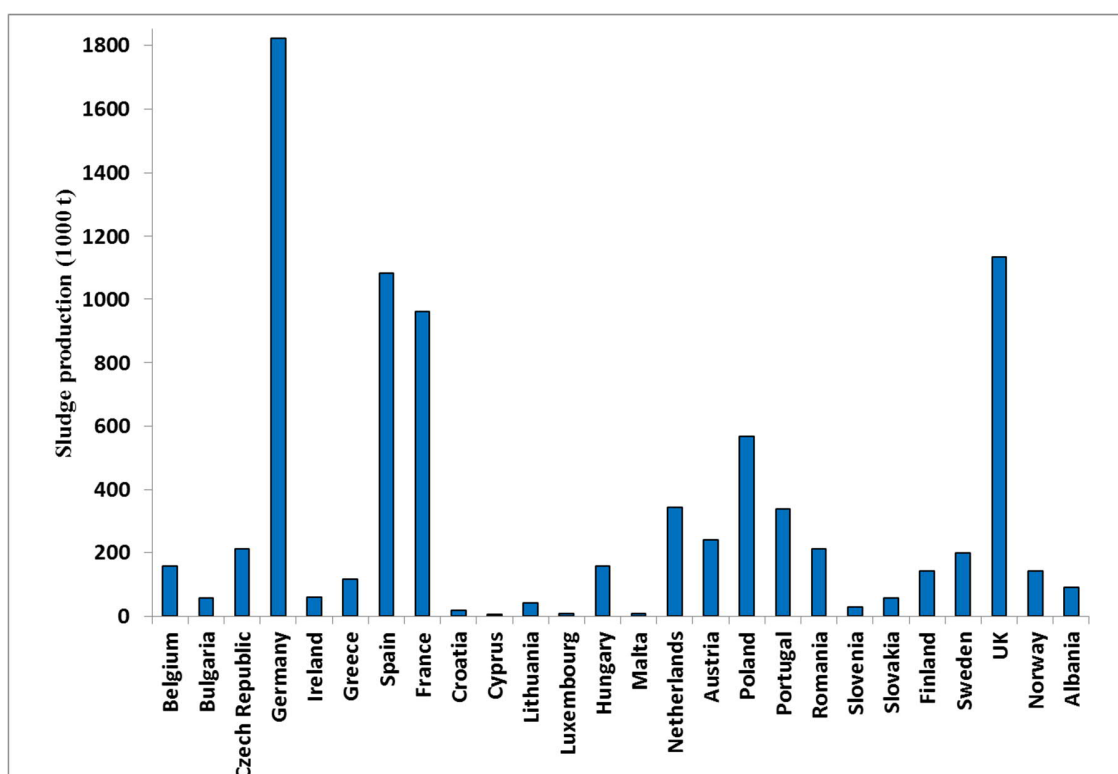
66 In addition to beneficial plant nutrients, sludge may contain a variety of inorganic and
67 organic substances, pharmaceuticals, and pathogens depending on the inputs of effluents in
68 the wastewater plants and types of digestion used in the process (Carrington, 2001; Urbaniak
69 et al., 2017; Hudcová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

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

84 **2. Production and disposal of sludge**

85 The alternatives for the disposal of sludge are agricultural use, composting, incineration, and
86 landfill (Epstein, 2003; EUROSTAT, 2018; Hudcová et al., 2019). In the past, agricultural
87 use (37% of the sludge produced in Europe) and landfill (40%) were considered the most

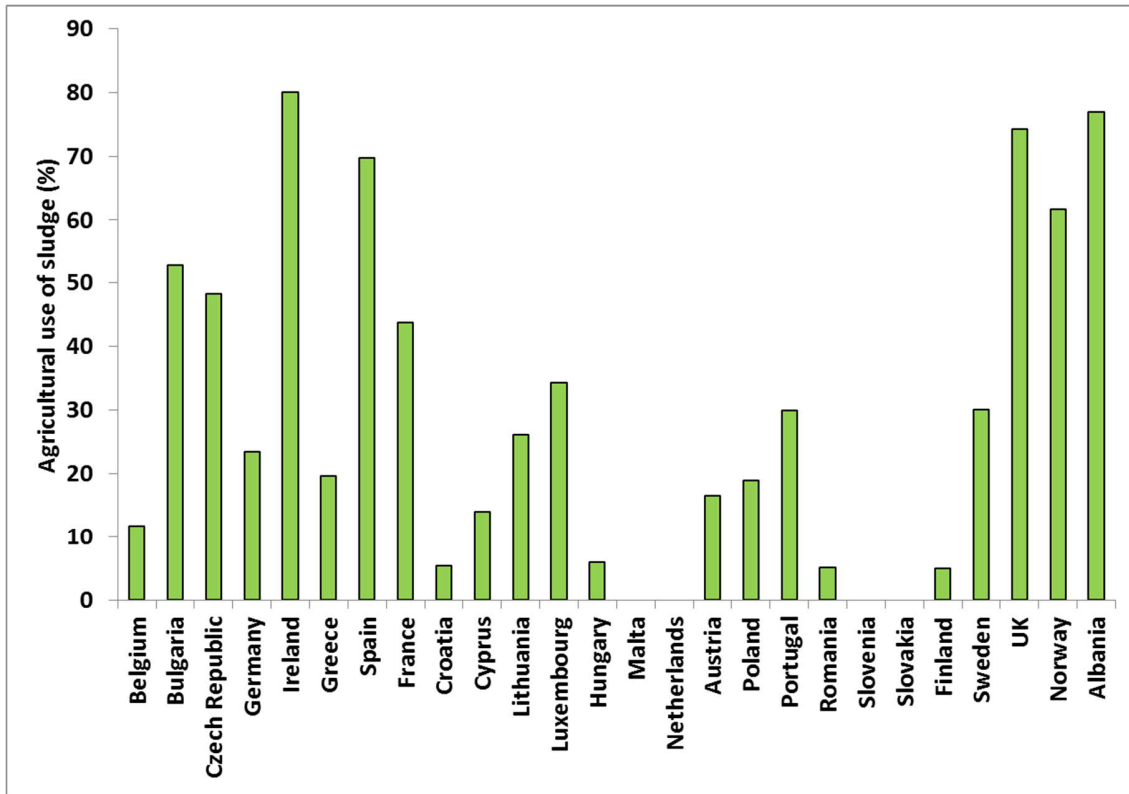
88 economical options for sludge disposal (Tchobanoglous and Burton, 1991; Fytli and
 89 Zabaniotou, 2008), followed by incineration (11%) and some other uses (12%) such as
 90 forestry and land reclamation (Fytli and Zabaniotou, 2008). However, incineration (39%)
 91 and agricultural use (25%) are considered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



98

99 **Figure 1**

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



102
103 **Figure 2**
104 Agricultural use (%) of sludge in the European Union during 2012 – 2015 (EUROSTAT,
105 2018).

106 3. Composition of sludge

107 3.1. Inorganic compounds in sludge

108 Sludge contains nutrients, heavy metals, and metalloids (Seleiman et al., 2012, 2013a,
109 2013b; Bai et al., 2017; Fijalkowski et al., 2017; Rehman et al., 2018; Wang et al., 2018),
110 some of which are essential macronutrients, such as nitrogen (N), phosphorus (P), potassium

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

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

132 to other nutrients (Xu et al., 2017), and meat-bone meal contains approximately 8.0% N,
133 4.4% P, 4.5% K, 1.5% Mg, and 10.0% Ca (Vamvuka et al., 2018).

134 **Table 1**
 135 Examples of chemical properties of sludge of different origins. In Seleiman et al. (2013a), sludge was based either purely on
 136 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					

137 DM, dry matter

138 **3.2. Organic compounds in sludge**

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

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

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

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

176 Advanced oxidation technologies are proved to be promising methods to decompose organic
177 pollutants from wastewater. Chemical, photochemical, sonochemical (ultrasound) and
178 electrochemical processes either individually or in different combinations based in
179 formation of very reactive, non-selective hydroxyl radicals initiate oxidative cascade leading
180 to mineralization of organic pollutants to CO₂ and H₂O (Vanraes et al. 2016, Deng and Zhao
181 2015, Segneanu et al. 2013).

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

189 **3.3. Pathogens in sludge**

190 Sludge may also contain pathogens, such as bacteria, viruses, protozoa, and eggs of parasitic
191 worms (Epstein, 2003; Yan et al., 2009; Urbaniak et al., 2017). Plant-based sludge could
192 contain plant pathogens, such as viruses, bacteria, fungi, and parasites, along with undesired
193 weed seeds (Carrington, 2001). Sludge of animal origin, such as from livestock production,
194 slaughterhouses, or meat processing industries, could contain antibiotic resistant bacteria
195 and viruses (Franke-Whittle and Insam, 2013). Slaughter wastes may also contain the prions
196 of bovine spongiform encephalopathy (Carrington, 2001; Franke-Whittle and Insam, 2013).
197 Typical concentrations of colony forming units of most common bacteria in untreated sludge
198 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
199 enteroviruses, 10^2 to 10^3 g⁻¹ of protozoa, and 5 to 10^3 g⁻¹ of helminth eggs (Carrington,
200 2001). To produce sludge for safe use as fertilizer in agriculture, the treatment must reduce
201 the number of *Salmonella spp.* and enteroviruses by at least four orders of magnitude as well
202 as destroy the viability of *Ascaris* and helminths' ova. In addition, the level of *Escherichia*
203 *coli* should not exceed 1000 colony forming units g⁻¹ DM sludge, and the level of

204 *Clostridium perfringens* spores should not exceed 3000 colony forming units g⁻¹ DM
205 (Strauch, 1998).

206 Pathogen level and diversity in wastewater vary temporally and geographically. Therefore,
207 their continuous detailed monitoring is impossible. Continuous detailed monitoring can be
208 replaced by the use of non-pathogenic surrogate organisms, which are sensitive indicators
209 of virulent human pathogens (Carrington, 2001). Organisms used in validation processes are
210 widespread in sludge and easy to maintain for validation tests *Escherichia coli* have proven
211 to be a suitable indicator by having similar characteristics to vegetative bacteria, for example
212 *Salmonella spp.*, *Shigella spp.*, *Listeria spp.*, and most viruses (Pike et al., 1988; Strauch,
213 1998).

214 Recycling sludge safely in agriculture requires environmentally and ecologically sustainable
215 technologies in efficiently remove hazardous organic pollutants, pharmaceuticals,
216 pathogens, and parasites from sludge. Common methods to stabilize and sanitize sludge in
217 WWTPs are biological anaerobic and aerobic digestion, heat treatment and alkaline
218 stabilization (Arthurson, et al. 2008; Goldfarb et al. 1999). Lime can be used to sanitize and
219 disinfect sludge, and remove different pathogens, particularly bacteria (Urbaniak et al.,
220 2017). Pathogenic microorganisms, which are found in sludge containing fecal and
221 vegetable material, have been listed in the European Commission's Evaluation of Sludge
222 Treatments for Pathogen Reduction – Final Report No: CO 5026/1 (Carrington, 2001).

223 **4. Sludge in agriculture and regulations regarding its use**

224 **4.1. Sludge on cropland**

225 Sludge added to cropland has many potentially beneficial impacts due to the improvement
226 of biological, chemical, and physical properties of soils, which may improve plant growth
227 and productivity (Beck et al., 1996; Seleiman et al., 2012, 2013b; Bai et al., 2017; Urbaniak
228 et al., 2017; Rehman et al., 2018; Hudcová et al., 2019). The high organic matter content
229 may improve the functioning of sludge as a soil conditioner by increasing soil water-holding
230 capacity and water infiltration, stabilizing soil temperature fluctuation, serving as a storage
231 of nutrients, and enhancing soil microbial activity (Jarausch-Wehrheim et al., 1999; Epstein,
232 2003; Samaras et al., 2008; Yan et al., 2009; Seleiman et al., 2013b).

233 On the other hand, many soil factors, such as pH, organic matter, aeration, cation exchange
234 capacity, water content, temperature, and elemental interactions, can affect the uptake of
235 elements from the soil (Epstein, 2003). Soil pH is considered the main factor affecting the
236 solubility of trace elements. For instance, the solubility of all essential trace elements except
237 molybdenum (Mo) and selenium (Se) is increased at low pH; hence their potential uptake
238 by plants increases. Depending on the environmental conditions, soil organic matter can
239 reduce or increase the plant availability of cationic trace elements, such as Cd, Cu, Ni, and
240 Zn, through chelate formation. High cation exchange capacity reduces the mobility of trace
241 elements, such as Cd, Cu, Ni, and Zn. Due to the high cation exchange capacity of clay
242 minerals, the binding of trace elements in clay soil is higher than in sandy soil (Epstein,
243 2003).

244 Similarly, soil organic matter has, on a mass basis, higher cation exchange capacity than the
245 mineral fraction of soil. The interactions between macronutrients and trace elements can
246 reduce the bioavailability of micronutrients in soil. For example, the interaction between the

247 phosphate and trace elements can form soluble or insoluble compounds depending on soil
248 pH (Epstein and Chaney, 1978). The less than optimal ratios of plant nutrient content in
249 sludge are generally complemented by adding commercial fertilizers with sludge to cropland
250 to balance the nutrition needed for each species (Yan et al., 2009) (Table 2).

251 **Table 2**

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

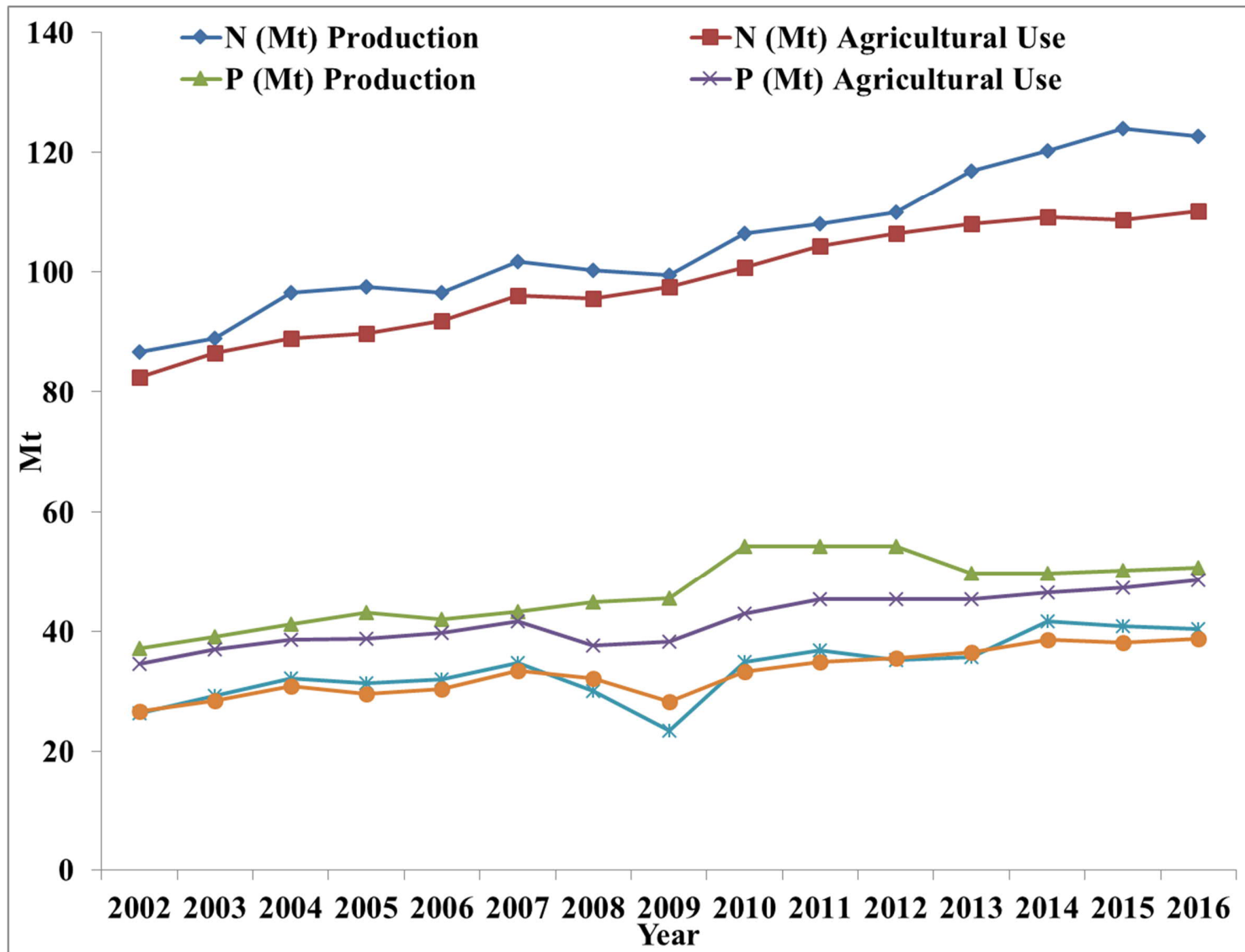
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254 According to Antoline et al. (2005) and Mendoza et al. (2006), sludge application increased
255 the ammonium-N content, cation exchange capacity, and organic carbon content in soil.
256 Unfortunately, sludge application also reduced soil pH and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

262 When applied as fertilizer, sludge increased the yield of alfalfa (*Medicago sativa* L.), wheat
263 (*Triticum aestivum* L.), faba bean (*Vicia faba* L.), zoysia grass (*Zoysia japonica* L.), and
264 biomass of maize (*Zea mays* L.), hemp (*Cannabis sativa* L.), oilseed rape (*Brassica napus*
265 L. ssp. *oleifera* [Moench.] Metzg.), willow (*Salix viminalis* L.), and sweet sorghum
266 (*Sorghum bicolor* L.) (Elsokkary and El-Keiy, 1988; Wang et al., 2008; Seleiman et al.,
267 2012, 2013a; Urbaniak et al., 2017; Rehmana et al., 2018; Zuo et al., 2019). However, it also
268 increased the heavy metal and metalloid content in the leaves and grains of barley (*Hordeum*
269 *vulgare* L.) (Antoline et al., 2005) and sorghum leaves (*Sorghum bicolor* L.) (Mendoza et
270 al., 2006), as well as the biomass of maize, hemp, oilseed rape (Seleiman et al., 2012, 2013a,
271 2013b), and sweet sorghum (Zuo et al., 2019), without any visual symptoms of heavy metal
272 toxicity.

273 Apart from improving soil structure and nutrient recycling, agricultural use of sludge also
274 has the potential to save energy compared with synthetic fertilizers. Almost 40% of the
275 commercial energy used in agriculture is consumed for manufacturing synthetic N fertilizers

276 due to the large energy input required for reducing N₂ to ammonia in the Haber–Bosch
277 process (Mudahar and Hignett, 1985; Hessel, 1992). The energy needed to produce synthetic
278 fertilizers including the packaging, transportation, and application to the cropland is about
279 78 MJ kg⁻¹ N, 17 MJ kg⁻¹ P, and 14 MJ kg⁻¹ K (Hessel, 1992; Elsayed and Mortimer, 2001;
280 Mikkola and Ahokas, 2009). Production of N fertilizers increased from 11.6 to 122.7 Mt and
281 synthetic P fertilizers from 10.0 to 50.8 Mt during the period 1961–2016 (IFASTAT, 2016;
282 FAOSTAT, 2012, 2019). During the period 2002–2016, the agricultural use of N fertilizers
283 increased from 82.5 to 110.2 Mt and the use of P fertilizers from 34.6 to 48.6 Mt (Figure 3)
284 (FAOSTAT, 2019).



286 **Figure 3**

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

288 (FAOSTAT, 2019).

289 **4.2. Main regulations related to use of sludge on cropland**

290 Sludge can contain, depending on the source and processing technique in WWTPs, a
291 quantitatively and qualitatively variable range of harmful compounds, pathogens, and
292 pharmaceuticals (Carrington, 2001; Clark and Smith 2011; Seleiman et al., 2013a; Al-
293 Gheethi et al., 2018; Hudcová et al., 2019). Therefore, its use on land is usually regulated.
294 The amount of heavy metals in sludge as well as in soil to which sludge is applied is strictly
295 regulated in the EU (Table 3). Council Directive 86/278/EEC of 12 June 1986 aimed to
296 protect the environment and prevent the harmful effects of adding sludge to cropland to
297 plants, animals, and humans (European Commission, 1986). The directive restricts: 1)
298 contamination of the soil by heavy metals and metalloids by limiting the contents of heavy
299 metals and metalloids in the soil to which sludge is applied, 2) the content of heavy metals
300 and metalloids in sludge, as well as 3) the amounts of heavy metals that can be annually
301 added to the soil (Table 3) and the maximum number of pathogens in sludge (European
302 Commission, 1986). Some EU countries have more restrictive requirements compared with
303 Directive 86/278/EEC and have approved the limits for heavy metal concentrations,
304 synthetic organic compounds, and microbial contamination (Hudcová et al., 2019). For
305 example, in the Netherlands the limits of Cd, Cu, Ni, Pb and Zn concentrations in sludge
306 should not exceed 1.2, 75, 30, 100, and 300 mg kg⁻¹, respectively (EUROSTAT, 2018;
307 Hudcová et al., 2019).

308 In the EU, specific requirements for organic compounds in sludge are not included in
309 Directive 86/278/EEC, but to reduce potential health risks several national regulations
310 include limitations concerning the allowable amounts of organic compounds in the sludge

311 applied to cropland. For example, the limits of PAHs and PCBs were 6.5 and 1.0 mg kg⁻¹
 312 DM sludge in Bulgaria, respectively, while the limits of DEHP, LASs, NPEs, and PAHs
 313 were 50, 300, 10, and 3 mg kg⁻¹ DM sludge in Denmark, respectively. In addition, the limits
 314 of adsorbed organic halogen compounds (AOX) and PCBs were 500 and 0.2 mg kg⁻¹ DM
 315 sludge in Germany, respectively (European Commission, 2000; Hudcová et al., 2019).

316 **Table 3**

317 European Union limits for heavy metals in sludge and soil to which sludge is applied
 318 (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
<i>Previous regulations(before 2015)</i>						
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
<i>Current regulations from 2015</i>						
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
<i>Proposed regulations 2025</i>						
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

319 DM, dry matter

320 In the USA, the Environmental Protection Agency's regulation 40 CFR Part 503 Standards
 321 for the Use or Disposal of Sewage Sludge (U.S. Federal Register, 1993) defines two sludge
 322 classes: Class A and Class B. Class A sludge has undergone composting, heat drying, and
 323 high-temperature aerobic digestion, which reduces pathogenic bacteria, enteric viruses, and
 324 viable helminths' ova to below detectable levels. Class A sludge can be used as a soil

325 amendment without imposing site and harvesting restrictions. Class B sludge may still
326 contain some pathogens, and thus its application is restricted for the harvestable crops,
327 animal grazing, and public access for a period of time after application (U.S. Federal
328 Register, 1993).

329 Sludge application is also limited in many European countries either as a maximum amount
330 $\text{ha}^{-1} \text{ year}^{-1}$ or by P-based agronomic rates. In the USA, a N-based application rate is used
331 and no specific limitations for organic contaminants exist (Harrison et al., 2006).

332 **5. Risk of contamination of food chain and the environment**

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

340 Even though sludge is a valuable source of nutrients, for example excessive P accumulation
341 in soil as a source of pollution for surface water and groundwater is of concern (Grobelak et
342 al., 2017). This can be caused by the non-optimal ratios of plant nutrients in sludge. Sludge
343 application at recommended rates based on the content of available soil P might result in an
344 accumulation of P in soil, which can increase the risk of eutrophication and an adverse
345 impact on water bodies through surface runoff, subsurface drainage water, and eroded soil.
346 To avoid leaching of P into the environment, wastewater treatment facilities have been

347 required to meet stricter effluent limits for P (Łuczkiewicz, 2006). Sludge produced in
348 WWTPs, where Fe, aluminum (Al), or Ca are used during pretreatment to reduce the soluble
349 P (to meet effluent limits), also has lower P available to plants (i.e., less than 25% of that in
350 triple superphosphate). Phosphate is strongly adsorbed to the surfaces of Fe and Al hydrous
351 oxides and calcium carbonate (Bastin et al., 1999). In addition, heat-dried sludge has low P
352 availability.

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

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

369 A high content of persistent and bioaccumulative organic contaminants in sludge is a concern
370 when sludge is used on cropland and in landfills, which may result in the spread of organic
371 pollutants into the environment and food chain. Many organic pollutants include a number
372 of congeners with different chemical quality and toxicities (Hudcová et al., 2019). The
373 concentrations of many of those “traditional” organic contaminants in sludge is regulated by
374 source control, but there exist still vast number of emerging potential compounds whose
375 toxicity and behavior in soil and water bodies have not been analyzed yet (Eriksson et al.
376 2008).

377 Temperature and time affect the degradation rate of LAS and NPE compounds in soil, which
378 must be considered when timing sludge storage and application to soil. For example, the
379 half-life of LAS at 22 °C was 7 days in comparison with 14 days at 13 °C (González et al.,
380 2010), and it took 56 days for NPE and 23 days for LAS to lose toxicity in the soil (Garrido-
381 Perez et al., 2008). Thus, LAS and NPE compounds can be loaded into soil via sludge and
382 transferred into surface water and groundwater. In the environment, NPEs degrade to
383 nonylphenol (NP) which is more persistent and toxic especially in aquatic environment (U.S.
384 Environmental Protection Agency, 2010).

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

388

389 Clarke and Smith (2011) ranked selected emerging organic compounds using an assessment
390 matrix approach with five properties: 1) environmental persistence (> 6 months), 2) human

391 toxicity, 3) bioaccumulation, 4) ecotoxicity, and 5) number and quality of international
392 studies of the contaminant. On the basis of the results, the organic compounds were
393 organized in decreasing priority as follows: perfluorinated chemicals (PFCs) >
394 polychlorinated alkanes (PCAs) and polychlorinated naphthalenes (PCNs) > organotins,
395 polybrominated diphenyl ethers, triclosan and triclocarban > benzothiazoles > antibiotics
396 and pharmaceuticals > synthetic musks > bisphenol A and phthalate acid esters and
397 quaternary ammonium compounds > steroids, and polydimethylsiloxanes.

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

402 Short-chain perfluorochemicals (PFCs), perfluorooctanoic acid (PFOA), perfluorooctane
403 sulfonates (PFOS), and polychlorinated alkanes (PCAs) have caused concern. All these
404 substances have been found in human blood (Olsen et al., 2003), human milk (Thomas et
405 al., 2006), and the environment (Campbell and McConnell, 1980; Giesy and Kannan, 2001).

406 PCAs are persistent and bioaccumulative and their mean concentration of sludge collected
407 from WWTPs in UK was much higher, 1800 mg kg⁻¹ DM in comparison to other persistent
408 organic pollutants (Stevens et al 2003). Short-chain PCAs and PFOAs are identified as
409 priority hazardous substances by European Water Framework directive and placed
410 restrictions on marketing and use (REACH Annex XVII).

411 Uptake of organic compounds by plants depends on the lipophilicity and water solubility of
412 the substance, the ambient temperature, as well as the concentration of organic compounds

413 in the soil and plants (Simonich and Hites, 1995). Organic compounds are taken up from a
414 soil solution by roots and translocated into the leaves. Some of these compounds can be
415 metabolized during the uptake and some intermediates can be translocated from the roots to
416 other plant parts, while others can remain in the plant roots. Volatile organic contaminants
417 can enter foliage through the wax-containing cuticle, which can bind and store lipophilic
418 organic compounds, as well as through the stomata (Duarte-Davidson and Jones, 1996).
419 Maize uptake of PFCs from nutrient medium was dependent on chain length of substances.
420 For instance, short-chain substances were taken up more actively than long-chain substances,
421 and translocated to shoot. Whereas higher concentration of long-chain substances, for
422 example PFOA and PFOS, accumulated in roots (Krippner et al. 2014). Wheat and
423 earthworms (*Eisenia fetida* L.) take up PFOS precursor perfluorooctane sulfonamide, which
424 is transformed to PFOS in both organisms and other derivatives in wheat (Zhao et al. 2018).
425 Organic compounds in soil treated with sludge can translocate and accumulate in grazing
426 livestock, depending on some factors such as livestock species, type of sludge applied to the
427 soil, and growing season. For example, PAHs could adhere to plant root surfaces and the
428 lighter molecular weight compounds might volatilize from polluted soil into the foliage.
429 Grazing livestock can ingest the organic compounds in three different ways: (a) grazing, (b)
430 adhering to soil and/or sludge with harvested forage, and (c) ingestion of the soil–sludge
431 mixture. For instance, up to 2% of a sheep’s diet is soil (Duarte-Davidson and Jones, 1996).
432 Nonpolar and persistent organic compounds can be stored and accumulated in fat tissues and
433 milk if not metabolized (Wild and Jones, 1992). Other organic compounds such as PCDD/Fs,
434 PCBs, and hexachlorobenzene are not metabolized (Geyer et al., 1987). In fact, the potential

435 accumulation of PCDD/Fs in livestock is a critical issue, since about two thirds of humans
436 get PCDD/Fs from animal fat – mostly dairy products and beef (Fürst et al., 1990; Beck et
437 al., 1992).

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

451

452 **6. Conclusions**

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

462 **Conflicts of Interest:** The authors declare no conflict of interest.

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