



## Use of sewage sludge in agricultural soils: Useful or harmful

Sezai Delibacak <sup>a,\*</sup>, Lyudmila Voronina <sup>b,c</sup>, Ekaterina Morachevskaya <sup>b</sup>

Ali Rıza Ongun<sup>a</sup>

<sup>a</sup> Ege University, Faculty of Agriculture, Department of Soil Science and Plant Nutrition, İzmir, Turkey

<sup>b</sup> Moscow State University, Faculty of Soil Science, Department of Agrochemistry and Plant Biochemistry, Moscow, Russian Federation

<sup>c</sup> Center for Strategic Planning and Management of Biomedical Health Risks, Moscow, 119121, Russian Federation

### Abstract

Sewage sludge is an important type of organic wastes among the various categories of solid waste. Organic matter resources in soils are relatively low and frequently require replenishment. Therefore, the use of sewage sludge in agricultural soils is a desirable method of their utilisation. The addition of sewage sludge to soils may be an inexpensive and effective alternative to the methods applied currently (mineral fertilisation, manure etc.). In spite of the undisputable advantages resulting from the application of sewage sludge in agriculture, it also involves some serious threats. Among those we should mention the presence of pathogens, heavy metals, and organic pollutants. In the current scenario of increasing global population, the generation of solid wastes like biosolids is bound to increase remarkably. Improper and unscientific disposal of biosolids results in several environmental issues such as surface and groundwater contamination, degradation of land, and food chain contamination. According to the principles of waste management hierarchy, agricultural recycling of biosolids will be a more environmentally preferred option over the traditional disposal methods. Utilizing the potential of biosolids to recycle valuable plant nutrients and as an effective soil amendment will not only help in sustainable management of this waste but also in minimizing the negatives associated with its traditional disposal. Every country must obey their regulations and legislations for managing their sewage sludge as a basic solution for use of sewage sludge in agricultural soil.

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### Introduction

Sewage sludge is an important type of organic wastes among the various categories of solid waste (Singh et al., 2014). In the present day, because of population growth, world of diminishing natural resources and energy crisis, the importance and need of developing a sustainable approach towards environmentally sound solid waste management cannot be ignored (Pappu et al., 2007). The improper disposal of solid wastes like biosolids and other biowastes pose a serious threat to the environmental quality leading to problems like groundwater contamination, degradation of soil quality, etc. Over the time, different approaches of safe biosolids disposal such as incineration, soil application, land filling (Kominko et al., 2017) and sea dumping have been explored (Sanchez Monedero et al., 2004). Disposal methods like land filling and ocean dumping have their own demerits due to scarcity of land, pollution problem and also don't lead to reuse of the beneficial constituents of biosolids (Wong, 1995; Singh and Agrawal, 2008). As a result, the United States and several European countries have banned the ocean dumping of biosolids since 1991 and 1998 (USEPA, 1999a,b; Zhidong and Wenjing, 2009).

\* Corresponding author.

Ege University, Faculty of Agriculture, Department of Soil Science and Plant Nutrition, 35100 Bornova, İzmir

Tel.: +90 232 3112653

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E-mail address: [sezai.delibacak@ege.edu.tr](mailto:sezai.delibacak@ege.edu.tr)

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Efficient municipal waste water treatment produces vast amounts of sludge. For example, in the countries located wholly or partly on the Baltic Sea watershed the amount of sewage sludge generated is about 3.5 million tonnes of dry solids annually. This is expected to increase to almost four million tonnes by 2020. Sludge management is an integral part of any modern municipal waste water treatment plant: it is important not to lose nutrients in the sludge, to make use of its material and energy, and to dispose of it efficiently and sustainably. During recent years, much effort has been put to efficient nutrient removal from the municipal waste waters in the Baltic Sea region. The aim is to reduce the eutrophication burden to the fragile Baltic Sea by fulfilling the relevant requirements: the newer EU member states on the eastern and southern shores of the Baltic Sea have started to implement the Urban Waste Water Directive (see chapter 12 for legislation) and also to strive for the stricter HELCOM recommendations for nutrient removal ([www.purebalticsea.eu](http://www.purebalticsea.eu)). Also, in the non-EU countries of the region, actions have been taken and are still on-going to improve the level of waste water treatment. The two main sludge fractions are primary and excess secondary sludge. If phosphorus is removed from the sewage by chemical precipitation, the amount of the sludge increases by the amount of chemicals used for the precipitation. The nutrients removed from waste water are contained in the sludge, which must be handled so that the nutrients are not released back to the watercourses, while the material and energy content of the sludge can be utilised. The large amounts of sewage sludge generated in waste water treatment plants provide numerous opportunities for beneficial use; for example, in power generation, soil improvement and even nutrient recycling. The possibilities of use are dependent on the quality and amount of the sludge in question, the processes used in a particular treatment plant, and the national legislation and policies. The sludge handling strategy of an individual waste water treatment plant is shaped, for example, according to: its location, transportation costs; the quality of incoming waters; nutrient removal technology used; legal restrictions concerning sludge disposal; the availability and price of conditioning agents; and the possibilities to outsource the treated sludge products. An accurate and unbiased assessment of the risks connected to the use of sewage sludge is needed. The question is how to deal with various chemical substances present in municipal sewage sludge originating from households and how to recycle the nutrients. The issue of hazardous chemicals of the sludge is currently under debate and regulatory issues are also open with the EU Sewage Sludge Directive currently being revised ([www.purebalticsea.eu](http://www.purebalticsea.eu)). There are international political dimensions to the sludge handling issue, not only in differing legislation, energy strategies and sludge handling costs, but also because the world's mineral resources of phosphorus are depleting. The question on nutrient recycling is emerging: according to some estimates, phosphorus resources may only be sufficient for the next 50 years. These resources are located mostly in northern Africa, China and the USA. Even though there have been some optimistic estimations on the global reserves, the dependency on a single country, Morocco, is expected to grow over the century. Thus, EU food security has to rely on imported phosphorus (Schróder et al., 2011). Municipal sewage sludge contains large amounts of precious phosphorus; however, the possibilities for recycling should be taken into account already when planning sludge management alternatives. Moreover, some sewage treatment methods do not allow the phosphorus to exist in an easily usable form in the sludge ([www.purebalticsea.eu](http://www.purebalticsea.eu)).

Land application of sewage sludge has a great incentive in view of its fertilizer and soil conditioning properties, unless it contains toxic substances. The heterogeneous nature of sewage sludge produced at different treatment plants and the variations between seasons necessitates knowledge of the chemical composition of sewage sludge prior to the land application. Characteristics of sewage sludge depend on the waste water treatment processes and sludge treatment. Generally sewage sludge is composed of organic compounds, macronutrients, a wide range of micronutrients, non-essential trace metals, organic micro pollutants and microorganisms (Kulling et al., 2001).

The practical and technical challenges of sludge handling are the following:

a) stabilising sludge is not inert and can have an unpleasant odour; b) reducing the water content and sludge volume to the minimum; c) utilising the energy potential when economically possible; d) reducing the amount of harmful micro-organisms if people, animals or plants are in contact with the sludge; e) recovering phosphorus for agriculture ([www.purebalticsea.eu](http://www.purebalticsea.eu)).

The macronutrients in sewage sludge serve as a good source of plant nutrients and the organic constituents provide beneficial soil conditioning properties (Logan and Harrison, 1995).

Chemical characterization of sewage sludge collected for over 2 years from eight cities of Indiana, USA, showed that sewage sludge contained approximately 50% organic matter and 1–4% inorganic carbon. Organic N and inorganic P constituted the majority of total N and P, respectively, of sludge. Organic and inorganic C, organic N and inorganic P, Ca and Mg were present in relatively constant concentrations in a given sludge throughout the sampling period. Inorganic N, organic P, K and all other metals were quite

variable during the entire period of study. The largest deviations were found for trace elements/heavy metals such as Cd, Zn, Cu, Ni and Pb (Sommers et al., 1976).

Bradford et al. (1975) studied trace element concentrations of sludges from certain metropolitan areas of southern California to evaluate their interactions with soils to determine their effects on plant growth. The total concentrations of trace elements in sludges were highly variable depending on the sources, which were related to different industries discharging effluents in the sewerage system. Concentrations of Cu, Zn, Ni, Co and Cd were consistently greater in saturation extracts obtained from sludges than those obtained from soils of any sampling sites. The maximum total concentrations of B, Cd, Ni, Cu, Pb and Zn showed positive correlations with concentrations of these elements found in soil solution.

## Effects of the sewage sludge application on soil properties

### Physico-chemical properties

Land application of the sewage sludge is becoming more popular due to the possibility of recycling valuable components such as organic matter, N, P and other plant nutrients (Martinez et al., 2002). Sewage sludge application to soil enables the recycling of nutrients and may eliminate the need for commercial fertilizers in cropland (Sommers, 1977). The fertility of soil increased over a long period of time, as sludges are organic fertilizers (Archie and Smith, 1981).

Unwise sludge amendment may, however, disturb the soil properties especially when it bears high concentrations of metals and toxic constituents. The soil physical conditions have been improved by sewage sludge application (Epstein, 1975, Table 2). An increase in soil pH has been reported in soils applied with municipal sewage sludge (Tsadilas et al., 1995). Lowering of soil pH is also reported (Epstein et al., 1976). The changes in soil pH have been correlated with the calcium carbonate content of sludge and acid production during sludge decomposition (Sommers, 1977). Soil pH consideration is especially important in view of trace metal abundance in sewage sludge. Maiti et al. (1992) characterized the sewage sludges of Calcutta, India, to assess their fertilization value. Sewage sludge pH was found to be neutral to slightly alkaline and to have a higher salt content in winter than in the monsoon season. The cation exchange capacity (CEC) was higher during monsoon. Exchangeable  $\text{Ca}^{2+}$  was the dominant cation followed by  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ , and  $\text{K}^+$ . The sludges were rich in organic carbon and available N.

A comparison of the data on physico-chemical characterization of selected sewage sludge from different countries collected during 1998–2002 clearly showed that pH may vary from an acidic to an alkaline range (Parkpain et al., 1998; Martinez et al., 2002; Nandakumar et al., 1998).

Organic matter added to the soil as sewage sludge composts improved the soil properties, such as bulk density, porosity and water holding capacity (Ramulu, 2002; Table 1).

Table 1. Effect of sewage sludge amendments on selected soil physical, chemical, and biological properties (Singh and Agrawal, 2008)

Properties	Effect	References
<b>Physical</b>		
pH	Decrease	Epstein (1975), Nielson et al. (1998)
	Increase	Tsadilas et al. (1995), Nielson et al. (1998)
Soil aggregate stability	Increase	Ojeda et al. (2003)
Bulk density	Decrease	Ramulu (2002), Ojeda et al. (2003)
Water holding capacity	Increase	Epstein (1975), Ramulu (2002)
Porosity	Increase	Ramulu (2002)
Erosion	Decrease	Ojeda et al. (2003)
Humus content	Increase	Kulling et al. (2001)
<b>Chemical</b>		
Toxic elements	Increase	Kulling et al. (2001), Lopez-Mosquera et al. (2000)
Soil organic carbon	Increase	Kladivko and Nelson (1979)
Electrical conductance	Increase	Martinez et al. (2002), Ramulu (2002)
N and P	Increase	Martinez et al. (2002), Sommers (1977), Walter et al. (2000)
Cation exchange capacity	Increase	Ramulu (2002), Soon (1981)
<b>Biological</b>		
Yeast population	Increase	Kulling et al. (2001)
Pathogenic organisms	Increase	Kulling et al. (2001), Ramulu (2002)
Aerobic bacteria	Increase	Kulling et al. (2001), Ramulu (2002)

A study conducted to determine the effect of 0.5% sewage sludge application to soil on water retention, hydraulic conductivity and aggregate stability showed that raw, as well as digested sludge, increased the total soil water retention capacity with the greatest increase in the raw sludge amended soil (Epstein,1975). Sludge addition in soil caused a significant increase in soil hydraulic conductivity after 27 days of incubation. Effect of sewage sludge amendments on selected soil physical, chemical, and biological properties was given on Table 1 (Singh and Agrawal, 2008).

The metal concentrations in the sewage sludge depend on several factors such as: i) sewage origin, ii) sewage treatment processes, and iii) sludge treatment processes (Hue and Ranjith, 1994). The bioavailability of the sludge borne metals to soil is further influenced by soil properties such as pH, redox potential (Eh), sesquioxide content and organic matter, as well as sludge application rate (Hue and Ranjith, 1994; Delibacak and Ogun, 2018).

Assuming that Zn, Cu and Ni behave similarly as pH varies, maintenance of a pH above 6.0 for grassland and 6.5 for arable soil to which the sewage sludge is applied, was recommended (Department of The Environment, 1981).

A study to evaluate the effect of pH on release of Zn, Cu and Ni from the sewage sludges showed that metal concentration released to the supernatant liquid increased as pH decreased below the threshold value, which was 5.8 for Zn, 6.3 for Ni and 4.5 for Cu loaded sludge (Adams and Sanders, 1984).

Hernandez et al. (1991) conducted a study to analyze the influence of sewage sludge application to a Calciorthid soil on the soil availability of macronutrients (N, P, and K) and

heavy metals (Fe, Cu, Zn, Mn, Ni, Cr, Cd, and Pb). Total N and extractable N and P contents increased in the sludge-amended soil, whereas the extractable K remained unaltered. Extractability of Fe, Cu, Mn, Zn and Pb increased due to sludge application as compared to the control. Relatively high rates of sludge application increased the cation exchange capacity, which helped to retain essential plant nutrients within the rooting zone due to additional cation binding sites (Soon, 1981).

Such responses, however, depend upon the sewage:soil ratio. The higher organic matter proportion in sludges decreased bulk density and increased the aggregate stability (Ojeda et al., 2003; Table 2). These improvements in soil physical properties increased water-holding capacity by promoting higher water retention in sludge-amended soils (Ojeda et al., 2003; Table 1).

Korboulewsky et al. (2002) studied the effects of the sewage sludge composts applied at the rates of 10, 30, and 90 t/ha fresh wt. on a vineyard in southeastern France to quantify in situ N mineralization and soil organic matter, and to evaluate environmental risks such as N and P leaching and accumulation of heavy metals in soil. It was found that soil organic matter increased at all of the treatment doses, but neither total nor available heavy metal concentrations increased. As the sewage sludge contained very low levels of heavy metals, mainly in nonextractable and nonexchangeable forms (Breslin, 1999), composting reduced the heavy metals availability in the raw material due to adsorption or complexing by humic substances. Mineral N increased in the first and in the second summers in the topsoil of amended plots. The risk of N leaching was very low, but P appeared to be the limiting factor at the recommended sludge amendment rate. Increase in P was significant in the top and in the subsoil of all of the treatment plots, with maximum increase at the highest rate of sludge application. At lower rates, no significant differences were observed. In soil, trace elements are distributed in various forms such as solid phases, free ions in soil solution, soluble organic mineral complexes, or adsorbed on colloidal particles. Sewage sludge addition to soils therefore could affect potential availability of heavy metals (Wang et al., 1997).

The solubility and consequently the mobility of metals added with sewage sludge are at least in part controlled by organic matter decomposition and the resultant soluble organic carriers of metals (Chaney and Ryan, 1993). Trace metal bioavailability is also dependent on the form of organic matter, i.e., soluble (fulvic acid) or insoluble (humic acid) (McBride, 1995). Insoluble organic matter inhibits the uptake of metals, which are tightly bound to organic matter and reducing the bioavailability. Soluble organic matters, however, increase the availability by forming soluble metal organic complexes (McBride, 1995).

The high organic matter content of acid soil led to the formation of complexes with Cu and thus impaired its uptake by plants. A contrasting trend was found with respect to metal concentrations in acid and alkaline soils. The concentrations of Zn, Cu, Pb and Ni in plants decreased with increase in sludge doses for acid soil. The study further suggested that soil type has a larger effect on the metal bioavailability than sludge doses (Morera et al., 2002).

## Microbial properties and soil enzymes

Land application of biosolids often results in significant changes in the structure, diversity, or richness of plant and animal communities at a given site, due directly, and indirectly, to resulting soil improvements and changing environmental conditions (USEPA, 1994). Few studies, however, have examined the soil microbial community in ecologically based studies, particularly in semi-arid grasslands (Dennis and Fresquez 1989; Pascual et al. 1999; Barbarick et al. 2004; García-Gil et al. 2004). Such research is crucial, considering that among other functions, microbial populations control decomposition and nutrient cycling in soils (Coleman and Crossley 1996), which in turn, control aboveground dynamics (Reynolds, 2018). Furthermore, a greater understanding of biosolids long-term effects on above- and belowground dynamics will lead to a greater understanding of ecosystem-level effects. Dennis and Fresquez (1989) found that cultivable fractions of the soil microbial community from a semi-arid grassland increased linearly with the biosolids application rate (22.5, 45, and 90 Mg ha<sup>-1</sup>), and they noted that improvements in soil fertility and site restoration were reflected in diversity and composition of the soil microbial community. García-Gil et al. (2004) reported increased microbial biomass, basal respiration, metabolic quotient, and enzymatic activities in another semi-arid soil, amended 9 or 36 months previously, with biosolids at approximately 36 Mg ha<sup>-1</sup>. Barbarick et al. (2004) examined microbial responses to biosolids in both semi-arid grassland (0 and 30 Mg ha<sup>-1</sup>) and shrubland (0 and 40 Mg ha<sup>-1</sup>) sites 6 years after biosolids surface application. At both sites, CO<sub>2</sub> evolution and actively metabolizing microbial biomass were greater in plots 6 years following biosolids application than in control (nonamended) plots. Pascual et al. (1999) examined microbiological and biochemical parameters of semi-arid soils 8 years after biosolids amendment (65 and 260 Mg ha<sup>-1</sup>). Compared to control plots, the amended plots exhibited greater total organic C, microbial biomass, basal respiration rate, and enzymatic activity even 8 years after biosolids incorporation. Enzymatic activities have often been estimated to establish the indices of soil fertility. Microorganisms as well as plants synthesize enzymes, which act as a biocatalyst of important reactions to produce essential compounds for both soil microorganisms and the plants. Soil enzymatic activities can be indirectly affected by heavy metals present in the sewage sludge (Kandeler et al., 2000). The effect of sewage sludge on biological activity may be used as indicator of soil pollution (Fließbach et al., 1994). According to some researches sewage sludge amendment increased the soil microbial activity, soil respiration and soil enzymes activities (Banerjee et al., 1997; Kayikcioglu and Delibacak, 2018). However, reduction in soil enzyme activities has also been reported at longer incubation period with high heavy metal availability (Fließbach et al., 1994). Effects of adding different doses (0, 100, 200 and 300 t/ha dry wt.) and C/N ratios (3:1, 6:1 and 9:1) of the sewage sludge on activities of b-glucosidase, alkaline phosphatase, arylsulphatase and urease in a clay loam soil at 25°C and 60% water holding capacity were studied by Kizilkaya and Bayrakli (2005). Nitrogen was added in the form of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> solution to the sludge to obtain different C/N ratios. Rapid and significant increase in the soil enzymatic activity has been noted at different doses and C/N ratios of the sewage sludge amendments as compared to unamended ones. Enzyme activities varied with differences in incubation period. Soils with the highest C/N ratio and sludge dose had the highest b-glucosidase activity. Alkaline phosphatase and aryl sulphatase showed an increment in their activity during the first 30 days of incubation followed by a pronounced decrease compared to unamended soil. Urease activity, however, showed an increase within 15 days, and thereafter activity declined. The highest activities of urease, alkaline phosphatase and arylsulphatase were observed in soil amended with a low C/N ratio and the highest dose of sludge (Kizilkaya and Bayrakli, 2005).

## Harmfull effects

Application of sewage sludge in agriculture or for soil reclamation is an interesting solution. It is more and more frequently used in practice due to the advantageous effect of sewage sludge on the properties of soils fertilised/reclaimed with its application. Apart from soil enrichment in nutrients (Fytili and Zabanitoutou, 2008), an addition of sewage sludge causes an increase in organic matter content in soil (Epstein, 2003). Organic matter resources in soils are relatively low and frequently require replenishment. Therefore, the use of sewage sludge in agriculture is a desirable method of their utilisation. The addition of sewage sludge to soils may thus be an inexpensive and effective alternative to the methods applied currently (mineral fertilisation, manure etc.). In spite of the undisputable advantages resulting from the application of sewage sludge in agriculture, it also involves some serious threats. Among those we should mention the presence of pathogens, heavy metals, and organic pollutants (Harrison et al., 2006; Oleszczuk, 2006a; Smith, 2009). Standards for maximum concentrations of pathogens in sewage sludge were given on Table 2.

Table 2. Standards for maximum concentrations of pathogens in sewage sludge (European Commission, 2009).

Country	Salmonella	Other pathogens
Poland	No occurrence	Faecal streptococci: < 100/g
France	8 MPN/10g DM	Enterovirus: 3 MPCN/10g of DM Helminths eggs: 3/10g of DM
Finland	Not detected in 25 g	Escherichia coli < 1000 cfu
Italy	1000 MPN/g DM	-
Luxembourg	-	Enterobacteria: 100/g no eggs of worm likely to be contagious
Hungary	-	Faecal coli and faecal streptococci decrease below 10% of original number
Poland	Sludge cannot be used in agriculture if contains salmonella	-

Proposed limit values on potentially toxic elements (PTE) in sewage sludge and in soil were presented in Table 3.

Table 3. Proposed limit values on potentially toxic elements (PTE) in sewage sludge and in soil (mg kg<sup>-1</sup> dw) (European Commission, 2009).

Metal	Sludge	Soil		
		5<pH<6	6<pH<7	pH>7
Cd	10	0.5	1	1.5
Cr	1000	50	75	100
Cu	1000	30	50	100
Hg	10	0.1	0.5	1
Ni	300	30	50	70
Pb	750	70	70	100
Zn	2500	100	150	200

The treatment of sewage sludge for agricultural purposes in Russia is regulated by two main documents, which allow to exclude to a certain extent the negative impact on the environment and human health. These are State All-Union Regulations R 17.4.3.07-2001 "Requirements for the composition and properties of sewage sludge when used as fertilizers" (Table 4), focusing on agrochemical and soil indicators, and Sanitary and epidemiological rules 2.1.7.573-96 "Hygienic requirements for the use of wastewater and its precipitation for irrigation and fertilizer" (Table 5), which determines the requirements for the quality of precipitation from a hygienic point of view.

Table 4. Permissible total content of heavy metals and arsenic in sewage sludge

Metals	Concentration, mg kg <sup>-1</sup> dry matter	
	1	2
Pb	250	500
Cd	15	30
Ni	200	400
Cr	500	1000
Zn	1750	3500
Cu	750	1500
Hg	7,5	15
As	10	20

Table 5. Sanitary-bacteriological and sanitary-parasitological indicators of sewage sludge

Indicator	Standard	
	1*	2**
E.coli group bacteria, cells g <sup>-1</sup> actual moisture	100	1000
Pathogenic microorganisms, including Salmonella, cells g <sup>-1</sup>	-	-
Geohelminth eggs and cysts of intestinal pathogenic protozoa, sample kg <sup>-1</sup>	-	-

1,2 - groups of sewage sludge

\*Sewage sludge of group 1 is used for all kinds of crops, except vegetables, mushrooms, green and strawberries.

\*\*Sewage sludge of group 2 is used for leguminous, cereals and industrial crops.

Sewage sludge of both groups is used in industrial floriculture, green building, forest and ornamental nurseries, for biological reclamation of disturbed lands and landfills.

Very rarely do urban sewerage systems transport only domestic sewage to the treatment plants. Industrial effluents and storm-water run off from roads and other paved areas are frequently discharged into the sewerage system. Thus sewage sludge may also contain many toxics in addition to organic material. Sewage sludge may also contain other harmful toxics such as detergents, various salts and pesticides due to effluents from municipal, industrial premises, toxic organics and hormone disruptors (Sommers et al., 1976). Organic pollutants are particularly important, due to its diversity. That diversity is related both with the method of toxic effect, and with diversified effects on living organisms (mutagenic effect, carcinogenic effect, endocrine disrupting effect) (Singh and Agrawal, 2008). With the development of analytical techniques, more and more often new potentially toxic organic compounds are identified in sewage sludge (Müller et al., 2006; Clarke and Smith, 2011; Davis et al., 2012).

Due to the multitude of those substances, and to the fact that their identification requires professional equipments, such contaminants are usually not the subject of routine chemical analyses. The lack of accurate information in this respect increases the risk involved in the application of sewage sludge. In this situation, biological tests may be helpful in the identification of potential threats. Biological tests permit not only measurable determination of a threat (toxic effect) but also take into account the possible interactions among the particular contaminants (antagonism/synergy effect). Moreover, the use of biological tests permits estimation of threats related with the presence of so far unidentified contaminants with potentially toxic effect. In this aspect, the estimation of phytotoxicity of sewage sludge is of particular importance due to its frequent utilisation for natural purposes. Moreover, plants are essential primary producers in the terrestrial ecosystem, whereas crop yield and quality are important success criteria in agriculture.

The application of phytotoxicity tests, therefore, permits not only to evaluate the applicability of sewage sludge for agricultural or soil reclamation purposes, but also to identify potential threats for the environment and for human health. Most of the studies concerned with the estimation of phytotoxicity of sewage sludge have been focused on the estimation of toxicity of the sewage sludge as such (Ramírez et al., 2008a;b; Hu and Yuan, 2012). Those studies, however, do not take into account other significant parameters, important in the assessment of the natural utilisation of sewage sludge. The toxicity of sewage sludge can also be significantly affected by the type of soil in which it is introduced (Domene et al., 2010), as well as by the kind of matrix under estimation (water extract or solid phase) (Domene et al., 2008). On the basis of studies conducted so far (Suchkova et al., 2010), it is also to be supposed that the species of plants grown can have a significant effect of the phytotoxicity of soil amended with sewage sludge, especially in the long-term approach. In spite of the importance of those problems, however, the literature lacks a comprehensive approach to those issues.

Oleszczuk et al. (2012) stated that their study aim was the estimation of changes in the phytotoxicity of soils amended with sewage sludge with relation to *Lepidium sativum*, *Sinapis alba* and *Sorghum saccharatum*. The study was realised in the system of a plot experiment for a period of 29 months. Samples for analyses were taken at the beginning of the experiment, and then after 5, 17 and 29 months. Two kinds of sewage sludge, with varying properties, were added to a sandy soil or a loamy soil at the dose of 90 t/ha. The addition of sewage sludge to the soils at the start of the experiment caused a significant reduction of both seed germination capacity and root length of the test plants, the toxic effect being distinctly related to the test plant species. With the passage of time the negative effect of sewage sludge weakened, the extent of its reduction depending both of the kind of sewage sludge applied and on the type of soil. Phytotoxicity of the soils amended with the sewage sludges was significantly lower at the end of the experiment than at the beginning. The species of the plants grown on the soils also had a significant effect on their phytotoxicity. The greatest reduction of toxicity was observed in the soil on which no plants were grown (sandy soil) and in the soil under a culture of willow (loamy soil). Solid phase of sewage sludge-amended soils was characterised by higher toxicity than their extracts. Most likely the reason of phytotoxicity was high doses of sewage sludge application.

A number of studies concerning the fate of organic and inorganic pollutants in soils amended with sewage sludge indicate that those compounds undergo a variety of processes (e.g. adsorption, desorption, bioformation, volatilisation, photodegradation, bioaccumulation, leaching and incorporation into humic substances structures -sequestration or bound residue formation-) (Hesselsøe et al., 2001; La Guardia et al., 2001; De Jonge et al., 2002). Those processes significantly determine the bioavailability of the pollutants (Alexander, 2000) and, indirectly, also their toxicity. In a study, after 29 months, in almost all treatments a significant decrease of phytotoxicity was observed in soils amended with sewage sludge in relation to the beginning of the experiment. Most probably that was a result of combination of all of the processes mentioned above. Moreover decreasing of the toxicity of the sewage sludge amended soil could be related

with quickly degraded labile compounds which very often occurs in sewage sludges and act as phytotoxins. The increase of phytotoxicity observed after 5 months in most of the experimental treatments was most probably related with remobilisation of pollutants, fairly frequently observed in soils amended with sewage sludge or composts (Oleszczuk, 2006b). Terry et al. (1979) and Rowell et al. (2001) stated that 26-42 % of the organic matter introduced together with the sludge underwent mineralisation very quickly. As a result of that process, formerly unavailable pollutants related with organic matter undergo remobilisation. An increase in the phytotoxicity after 5 months was most probably related to the fact that the organic contaminants, initially adsorbed to the sewage sludges/soil mixture, were temporarily less available. As a result of organic matter mineralization, the strength of these bonds could weaken, and hence, there was an increase in the bioavailability of pollutants which had not been bioavailable earlier (Oleszczuk, 2006b).

It is commonly accepted that toxicity largely depends on the water solubility of pollutants. However, studies on the sewage sludge phytotoxicity concentrate mainly on the analysis of water extracts (Wong et al., 2001; Fuentes et al., 2006; Mantis et al., 2005). The application of water extracts provides important information; however, they do not give a fully comprehensive description of the toxicity of sewage sludges. The results obtained in a study showed clearly that the analysis of extracts is not sufficient for the full characterisation of risks related with sewage sludge. This is also supported by earlier studies conducted with relation of other organisms and environmental matrices (Chial and Persoone, 2003; Ramirez et al., 2008a).

### Organic pollutants

Sewage sludges can contain significant concentrations of toxic heavy metals and organic pollutants (Guo et al., 2009). The sources of these pollutants in the waste waters are discharges from domestic and industrial facilities and atmospheric deposition as well as urban runoff (Blanchard et al., 2001; Harrison et al., 2006; Guo et al., 2009). Hence, these kinds of pollutants are concentrated in the sewage sludge during waste water treatment. This issue is of great risk to human health because transfer pathways of these pollutants into the human food-chain might be food crops or grazing livestock which eat contaminated feed grown on sludge-amended soil (McLachlan et al., 1996). Among these organic pollutants, polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental contaminants that originate from different emission sources, like the incomplete combustion of fossil fuels, industrial processes or the use of motor vehicles (Ozcan et al., 2013).

According to the data of Russian studies of the urban sewage sludge of Moscow, there are more than 200 polluting organic substances of anthropogenic nature, belonging to different groups of chemical compounds: acyclic saturated hydrocarbon, unsaturated hydrocarbons, aromatic cyclic hydrocarbons, polycyclic aromatic hydrocarbons, oxygen-containing compounds. Among them, 1.16% by weight belonged to toxic aromatic hydrocarbons, of which toluene was found in concentrations on average up to 10 times higher than MPC (maximum permitted concentrations) for soil. Also found naphthalene, methylnaphthalene, fluorene, phenanthrene, anthracene, pyrene and fluoranthene, belonging to the highly toxic group of polycyclic aromatic hydrocarbons (Kasatkov et al., 2017).

Concerning organics, the following are considered of primary importance for EU as limits are to be set in the revision of Sewage Sludge Directive: AOX (sum of halogenated organic compounds) linear alkylbenzene sulphonates (LAS) di(2-ethylhexyl)phthalate (DEHP) NPE (nonylphenole and nonylphenole ethoxylates with 1 or 2 ethoxy groups) polycyclic aromatic hydrocarbons (PAHs) polychlorinated biphenyls (PCBs) polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs). Limit values for concentrations of organic compounds in sludge of different countries and as suggested in the 3rd draft of the Working paper on sludge for EU (Langenkamp et al., 2001) were presented on Table 4.

Table 4. Limit values for concentrations (mg kg<sup>-1</sup> dw) of organic compounds in sludge of different countries and as suggested in the 3rd draft of the "Working paper on sludge" for EU (Langenkamp et al., 2001).

	AOX	DEHP	LAS	NP/NPE	PAH	PCB	PCDD/F, ng TEq/kg dm
EU	500	100	2600	50	6 <sup>1</sup>	0.8 <sup>2</sup>	100
Denmark	-	50	1300	10	3 <sup>1</sup>	-	-
Sweden	-	-	-	50	3 <sup>3</sup>	0.4 <sup>4</sup>	-
Lower Austria	500	-	-	-	-	0.2 <sup>5</sup>	100
Germany	500	-	-	-	-	0.2 <sup>5</sup>	100

<sup>1</sup> Sum of acenaphthene, phenanthrene, fluorine, fluoranthene, pyrene, benzo(b+j+k) fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, indeno(1,2,3-c,d)pyrene

<sup>2</sup> Sum of 6 congeners PCB 28, 52, 101, 138, 153, 180

<sup>3</sup> Sum of 6 compounds

<sup>4</sup> Sum of 7 congeners

<sup>5</sup> Each of the six congeners PCB 28, 52, 101, 138, 153, 180.

## Polycyclic aromatic hydrocarbons (PAHs) contents and sources

Polycyclic aromatic hydrocarbons are one of the most important groups of ecotoxicants due to their high toxicity, considerable stability in the environment, a cumulative effect. They are chemically inert and are hardly affected by the action of acids and oxidizing agents. Certain members of the PAH class have been listed as priority pollutants by Environmental Protection Agencies of the United States, Europe and China. The PAHs often detected in sewage sludge are mainly naphthalene, phenanthrene, anthracene, fluorene, acenaphthene, acenaphthylene, 1,2benzanthracene, benzo(a)pyrene, benzo(b) fluoranthene, benzo(g,h,i)perylene, benzo(k) fluoranthene, fluoranthene, indeno(1,2,3-cd)pyrene and pyrene (Zhai et al., 2011).

The origin of PAHs in sewage can be from combustion of petroleum, kerosene, grass, coal, and wood. Concentration ratios of some PAHs are often used to estimate the sources of PAHs, i.e., combustion and petroleum sources (Soclo et al., 2000). Using molecular mass of 178 as an indicator, anthracene to (anthracenesphenanthrene) ratio with  $<0.10$  usually represents the petroleum sources PAHs, while the ratio with  $>0.10$  is often considered as an indication of combustion sources (Budzinski, 1997). Yunker et al. (2002) reported that the ratio of (benz[a]anthracene)/(benz[a]anthraceneschrysene) lower than 0.20 implies a petroleum source; the ratio from 0.20 to 0.35 means either petroleum or combustion sources; and the ratio higher than 0.35 indicates combustion sources.

The study of samples collected at a waste-water treatment plant in Shanghai (China) indicates that PAHs release regularly according to temperature changes in the process of sewage sludge incineration treatment (Zhang et al., 2016). Over 90% of total PAHs in sewage sludge are released at the temperature of 300–750°C. The transformation of naphthalene to indeno (1, 2, 3cd) pyrene may be related to the temperature of the treatment system. It has been shown that the output rate of transformation reactions for indeno (1, 2, 3-cd) pyrene is 94% at 300°C.

The source of wastewater, wastewater treatment methods and different treatment process affect the PAH concentrations in sludge. The PAHs concentrations varied strongly between the sludges from primary and secondary treatment processes: the sum of PAHs in the primary sludge were 1.5973 and 0.3402 mg/kg, in the secondary sludge they were 8.7884 and 2.0185mg/kg respectively (Zhai et al., 2011). An enrichment of PAHs over time can be explained by the longer retention time of secondary sludge in comparison with primary sludge.

Previous studies reported a large range in the total concentration of PAHs in sewage sludge from Asian, African, and European countries, varying from below detection to 33,000 ng/g dry weight (Cai et al. 2007; Man et al. 2016; Poluszyńska et al. 2017).

According to Turkish regulation, concentration of total PAHs (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene Indeno[1,2,3-c,d]pyrene, dibenzo[a,h]anthracene, benzo[g,h,i]perylene, benzo[j]fluoranthene) should be  $<6 \text{ mg kg}^{-1}$  dry matter. According to EU regulation, concentration of total PAHs (acenaphthene, phenanthrene, fluorene, plouranthene, pyrene, benzo[b;j;k]fluoranthene, benzo[a]pyrene, benzo[g,h,i]perylene, indeno[1,2,3-c,d]pyrene) should be  $<6 \text{ mg kg}^{-1}$  dry matter. If the ratio is  $>0.50$ , the combustion of kerosene, grass, coal, and wood results in the PAH contamination. If fluoranthene/(fluoranthenespyrene) ratio for sludge was  $<0.50$ , which means the origin of PAHs was petroleum combustion. While this ratio for sludge was  $>0.50$ , indicating possible source of PAHs was combustion of kerosene, grass, coal, and wood (Ozcan et al., 2013).

Russian legislation in the field of health and environmental protection provides the standards only for the content of benzo (a) pyrene -0,02 mg kg<sup>-1</sup> (SanPiN GN 2.1.7.2041-06).

## Polychlorinated biphenyls (PCBs) contents

Although sewage sludge improves soil properties, numerous studies have demonstrated that they also contain toxic organic compounds, such as polychlorinated biphenyls (PCBs) (McLachlan et al. 1996; Urbaniak et al. 2014; Wyrwicka et al. 2014), PCBs are found in the natural environment because of human activities (Borja et al., 2011). Approximately 30% (i.e., 10 million tonnes) of the total worldwide production of PCBs has entered the environment (Benabdallah El-Hadj et al., 2007). The presence of these compounds in sewage sludge significantly decreases the potential for the sludge to be used in agriculture. According to proposed changes to EU Directive 1986/278/EEC, the polychlorinated biphenyl (PCB) concentration (expressed as the sum of the concentrations of the indicator congeners, PCBs 28, 52, 101, 118, 138, 153, and 180) should not exceed 0.8 mg/kg dry matter (DM) in sewage sludge used as a crop fertilizer. Many researches confirm the presence of PCBs in sewage sludge and organic waste, which should be utilized (Blanchard et al., 2004). Two biological methods are generally used: composting and the anaerobic

digestion. Rosinska and Dabrowska obtained decrease of PCB content over 50% during anaerobic digestion of sludge (Rosinska and Dabrowska, 2014). Despite the fact that PCBs show a strong affinity to organic matter which predispose them to be stored in the sediments (Gdaniec-Pietryka et al., 2013) and surface layer of the soil, it has been also shown that they can be transferred deeper into the soil profile (Bi et al., 2002; Kobasić et al., 2008; Zhang et al., 2011). Hence, the continued use of such contaminated sludge for agricultural purposes can present problems associated with the risk of soil, subsurface and groundwater contamination (Bi et al., 2002). This also concerns nutrients, which despite being valuable from the perspective of agriculture, their combined application with sewage sludge may lead to health and environmental risks associated with subsurface and groundwater pollution (Kang et al., 2011).

The Turkish and EU regulations describes that levels of PCBs (sum of PCB 28, 52, 101, 118, 138, 153, and 180) in sewage sludges should be  $<0.8 \text{ mg kg}^{-1}$  (dry weight) in order to be used in agriculture (Ozcan et al., 2013). Ozcan et al. (2013) indicated that the PCB concentrations of all investigated sewage sludges in their study were less than the limit value, it was found that use of them for agricultural purposes was suitable.

## Conclusions

Sewage sludge to be utilised in agriculture must be subjected to comprehensive evaluation comprising not only the determination of the basic physicochemical properties, content of pollutants or pathogenic bacteria, but also of the ecotoxicological properties. i) phytotoxicity of sewage sludge and its changes overtime are significantly determined by the soil type. In this case, the soil type is one of the most important factors regulating the phytotoxicity of sewage sludge, especially in the long-term aspect; ii) with the passage of time the phytotoxicity of soils amended with sewage sludge undergoes a change, not always in a direction causing a lowering of their toxicity. The extent of particular changes depends both on the properties of the soils and on the kind of sewage sludge, and for various kinds of sludge, it can have different direction in a single soil; iii) the extent of changes in the toxic effect on the test plants is related to the species of the plant under cultivation; iv) in spite of similar initial toxicity displayed by various kinds of sewage sludge in relation to particular plants, further changes of phytotoxicity may vary with relation to the sewage sludge and the type of soil; v) the intensity of toxicity towards various plants depends not only on the kind of sewage sludge but also on the soil type; vi) proper selection of the conditions of management sludge-amended soil can be conducive to a reduction of phytotoxicity; and vii) estimation of extracts is not sufficient for the full characterisation of the risk involved in the utilisation of sewage sludge in agriculture.

On the other hand, biosolids is a good source organic matter as well as plant macro and micro-nutrients and in future can be substituted for expensive inorganic fertilization. Addition of treated biosolids to soil has been found to be beneficial to soil health, enriching soil with essential nutrient elements as also increasing the pH of the soil. Further, soil amendments with biosolids has been reportedly effective in increasing a number of agro morphological attributes as well as yield in different crop species. However, use of biosolids for commercial agriculture has to be done cautiously. It is common knowledge that biosolids often contains toxic metal residues (heavy metals), as well as toxic organic residues, indiscriminate use of it can be detrimental to the productivity of the soil as well as cause harm to the food chain. Moreover the character of the biosolids changes over time and hence stringent and periodic monitoring of biosolids for agricultural use should be done. In general, a potential waste management depends on several tiers like disposal, recovery, recycle, reuse and prevention. This hierarchy is also suitable for managing biosolids. It is evident that implication of biosolids induce agricultural productivity to a certain level; but the application of this waste in major food generation and supply chain is still needs more study.

International as well as national guidelines on the carrying capacity of toxicants in biosolids composts should be set and stringent monitoring of such guidelines should be enforced in agricultural use of biosolids. Attempts for commercial production of biosolids based fertilizer and soil health amendments can be setup and can be used to boost agricultural production, and minimizing the dependency on inorganic fertilization. This will further reduce the carbon footprint, as most inorganic fertilizers are manufactured at high energy costs. Biosolids can be used for regaining soil fertility in agricultural lands deemed infertile by prolonged and indiscriminate use of synthetic inorganic fertilizers. In such conditions, depletion of soil nutrients as well as decreased soil pH can be rejuvenated with the rational use of biosolids. Owing to high nutrient efficiency and its importance in agriculture and soil fertility, stringent and persistent monitoring of biosolids and its products should be done before introduction in agricultural systems.

In the current scenario of increasing global population, the generation of solid wastes like biosolids is bound to increase remarkably. Improper and unscientific disposal of biosolids results in several environmental issues such as surface and groundwater contamination, degradation of land, and food chain contamination.

Hence, according to the principles of waste management hierarchy, agricultural recycling of biosolids will be a more environmentally preferred option over the traditional disposal methods. Utilizing the potential of biosolids to recycle valuable plant nutrients and as an effective soil amendment will not only help in sustainable management of this waste but also in minimizing the negatives associated with its traditional disposal. Every country must obey their regulations and legislations for managing their sewage sludge.

Researchers around the world are looking for the most optimal set of substances for priority analysis. The knowledge of which will be used to find the best path to deal with the problem of rational and, necessarily, environmentally-safe way of disposal of sewage sludge. Perhaps more fundamental research using new modern chemical methods for the analysis of hazardous organic compounds can tighten the control on the qualitative composition of sewage sludge used in agriculture.

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