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The presence of contaminations in sewage sludge – The current situation

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ABSTRACT

Sewage sludge/biosolids are by-wastes of municipal and industrial wastewater treatment. As sources of nutrients (C, N, P) they are widely used in intensive farming where large supplementation of organic matter to maintain fertility and enhance crop yields is needed. However, according to the report of European Commission published in 2010, only 39% of produced sewage sludge is recycled into agriculture in the European Union. This situation occurs mainly due to the fact, that the sewage sludge may contain a dangerous volume of different contaminants. For over decades, a great deal of attention has been focused on total concentration of few heavy metals and pathogenic bacteria *Salmonella* and *Escherichia coli*. The Sewage Sludge Directive (86/278/EEC) regulates the allowable limits of Zn, Cu, Ni, Pb, Cd, Cr and Hg and pathogens and allows for recovery of sludge on land under defined sanitary and environmentally sound conditions. In this paper, a review on quality of sewage sludge based on the publications after 2010 has been presented. Nowadays there are several papers focusing on new serious threats to human health and ecosystem occurring in sewage sludge – both chemicals (such as toxic trace elements – Se, Ag, Ti; nanoparticles; polyaromatic hydrocarbons; polychlorinated biphenyl; perfluorinated surfactants, polycyclic musks, siloxanes, pesticides, phenols, sweeteners, personal care products, pharmaceuticals, benzotriazoles) and biological traits (*Legionella, Yersinia, Escherichia coli* O157:H7).

1. Introduction

Sewage sludge can be defined as the solid or semi-solid residue left over after the treatment of wastewater. In literature it can be defined as by-product, yet it shall be treated as a waste in the process of wastewater treatment. Sewage sludge may be used as a source of energy (anaerobic digestion, thermal treatment), treated and used on land as a fertilizer and soil conditioner, or may even be used as a source to extract valuable compounds (phosphorous recovery). A significant number of wastewater treatment plants (WWTPs) compost dewatered sewage sludge under aerobic conditions with green wastes or other bulking agents or dry it in heat drying facilities up to 95% dry mass for use as fertilizer or fuel.

In most developed countries particular attention is drawn on proper treatment of sewage sludge to improve the quality and safe use on land. United States Environmental Protection Agency (US EPA) defines biosolids as treated sewage sludge that meets the suitable levels of pollutants or pathogen and is used as fertilizer for landscape application (USEPA, 2009). Wastewater sludge as a complex heterogeneous mixture of micro-organisms, undigested organics as cellulose, plant residues, oils, or fecal material, inorganic material, sand is a resource of organic matter, nitrogen, phosphorous, micronutrients and even heavy metals, bio-fuel, hydrogen, syngas, bio-oil, bio-diesel, bio-plastics, bio-pesticides, proteins, enzymes, bio-fertilizers or voilatile-acids (Tyagi and Lo, 2013). Currently the main trends in development of sustainable human communities include the investigation of the best strategies of the recycling of those precious substances (LeBlanc et al., 2009). However, taking into consideration standards set for waste which are reintroduced in natural systems, precautionary aspects shall be considered, especially on the limit values (quality criteria) for potential contaminants and pollutants dangerous to human health and the environment.

In Europe, the Sewage Sludge Directive (86/278/EEC) (SSD), the one of the oldest obligatory directive was set up to encourage the use of sewage sludge in agriculture and to regulate its use in such a

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way as to prevent harmful effects on environment by limiting the possible transfer of heavy metals and pathogens. Generally the Directive had the positive effect of improving source control measures in order to ensure a good quality of sludge, though currently it is considered as out-of-date and has been earmarked by the Commission as a candidate for revision for around 10 years (Environment, 2014). According to the report of European Commission published in 2010, only 39% of sewage sludge is recycled into agriculture in the EU due to increasing leaching of contaminants to water and soil, odors and greenhouse gas emissions (CH_4 and CO_2). Large variations are noted for sludge used on land in the Member States, ranging from none (Nederland, Switzerland) to over 50% (Norway, Great Britain, France). Coalition agreement of the federal government of Germany in November 2013 concluded: "*We will face out the direct use of sewage sludge as a fertilizer on land and promote the recycling of phosphorus and other nutrients*" (Bergs, 2015). At the other high-income countries, like USA, Canada, Australia, New Zealand treated biosolids are widely used on soils, however incineration has been suggested as a promising alternative of final sewage sludge disposal. Nevertheless, in less developed countries land application of treated sewage sludge is growing alternative for landfilling.

Hence the major question is what kind of contaminants can be found nowadays in sewage sludge? There are several papers focusing on new serious threats to human health and ecosystem occurring in sewage sludge – both chemicals (polyaromatic hydrocarbons (PAH), hydrocarbons; polychlorinated biphenyl (PCB), Perfluorinated Surfactants (PFCs), Personal Care Products (PCPs), Pharmaceuticals (PhCs), Benzotriazoles) and biologicals (*Legionella, Yersinia, Escherichia coli* O157:H7). However only some countries, e.g. Sweden initiated a program to systematically sample, analyze and bank sewage sludge (Olofsson et al., 2012). In the present paper we clearly demonstrate the necessity of introduction of monitoring program for emerging pollutants. Therefore a review on quality of sewage sludge based on the publications after 2010 has been presented.

2. Possible strategies for sewage sludge management

Sewage sludge as a waste (by-product) from the wastewater treatment process for many years has been mainly utilized by landfarming or/and landfilling. Due to the increasing content of toxic pollutants in many countries the sewage sludge has been found as hazardous waste and often incinerated. Sewage sludge (after meet the requirements of SSD in terms of limit values) is widely used as a fertilizer. However, the Member States significantly vary in the amount of generated sludge used in agriculture ranging from none to well over 50%.

From environmental point of view, agricultural use of sewage sludge is preferable because the organic and inorganic nutrients are recovered. Yet, nowadays, alternative treatment and disposal processes are selected and proposed (Fig. 1). As a result of conversion, sewage sludge loses its original properties and then becomes useful in the form of other products. Hence, a by-product can be obtained from biowastes. However, the change of waste into a product is also connected with a change of legislation. For instance, compost produced from sludge must meet the legal requirements for organic fertilizer.

Moreover, new technologies often require introduction of eco-innovative solution and holistic approach. OECD defined innovation as "the implementation of a new or significantly improved product (good or service), or process, a new marketing method, or a new organizational method in business practices, workplace organization or external relations" (Marchal et al., 2011).

3. Metallic trace elements

Inorganic contaminants are not biodegradable, and therefore can accumulate in the soil and enter the food chain and bioconcentrate in the environment. Historically, the most important are heavy metals, classified as chemical elements having a specific gravity higher than 4.5 g cm^{-3} (Kabata-Pendias, 2010). The most commonly used with this respect are Cr, Mn, Fe, Co, Ni, Cu, Zn, Hg, Cd, Pb, Sn, Mo, V. Though, likewise important from the standpoint of toxicity are metalloids like As, Se or non-metals and light metals as Al. Hence, a term metallic trace elements is more and more often used. Main source of heavy metals in sewage sludge are industrial wastewater and surface runoff. The total content varies within wide limits (from 0.5 to 2% of dry sludge). Taking into consideration the quantity of individual element it can be lined up as follows: Zn > Cu > Cr > Ni > Pb > Cd or Zn > Cr > Pb > Cu > Ni > Cd (Wilk and Gworek, 2009).

Many Member States have decided to implement stricter limit values (Table 1) than those stipulated by the Directive 86/278/EEC (Environment, 2014). This situation was noted particularly for mercury, but also for cadmium and nickel. In the case of zinc usually limit values close to the maximum allowed by the Directive have been adopted.

As shown by numerous studies, the total amount of metals in the sludge regulated by Directive 86/278/EEC is reduced, as indicated by the long-term analysis in Germany (Table 2). Since 1977, the largest decrease has been observed in the case cadmium, chrome and mercury – 95.4, 94.8 and 89.6%, respectively. It is noteworthy that the smallest divergence was noted for copper – since 1977 the content of this element has been reduced only by 22%. The content of heavy metals observed after 2000 year was lower comparing to the limit values proposed by Directive 86/278/EEC.

The total content of heavy metals is not a reliable indicator to assess their availability for living organisms, and thus the intrinsic toxicity. Such an assessment can be made by determining the amount of metal ions bound by the individual components (fractions) using sequence analysis. Lasheen and Ammar (2009) showed that Mn, Ni and Zn were most present in the exchangeable, carbonate and Fe/Mn-oxide forms as the most mobile fractions, while Cd, Cu, Cr and Fe were major in the organic and sulfide (exhibiting some degree of mobility), and the residual form (inert phase) which, corresponds to less mobilization. Different methods of stabilization alter both total concentration and the bioavailability of metal ions. The same authors confirmed that the use of cement kiln dust significantly reduced the availability of metals by chemical modification of their chemical speciation into less available forms. Dąbrowska and Rosińska (2012) did not observe accumulation of mobile fractions (exchangeable and carbonate) as an effect of thermophilic digestion of sewage sludge except for nickel. The highest increase of Zn, Cu, Cd and Cr concentration was observed in the form of organic-sulfide fraction, whereas in the case of Pb, the residual fraction noted the highest increase. For Ni both organic-sulfide and exchangeable–carbonate fractions were enriched. Smith (2009) in his critical review demonstrated the reduced bioavailability and crop uptake of metals from composted biosolids comparing to other types of sewage sludge. Furthermore, the use the earthworms affected metal speciation in vermicomposted sludge (Y. Zhang et al., 2008). In turn, Xiao et al. (2015) noted that after combustion process of pelletized municipal sewage sludge (MSS), the bioavailable heavy metal fractions (acid soluble/exchangeable, reducible and oxidizable fractions) were mostly transformed into the very stable heavy metal fractions (residual fractions). Healy et al. (2016) analysed metal concentrations in sewage sludge (SS) treated by thermal drying, lime stabilization, or