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REVIEW PAPER

CHEMICALS IN WASTEWATER AND SEWAGE SLUDGE – AN UNDERESTIMATED HEALTH AND ENVIRONMENTAL THREAT*

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Abstract

The objective of the work is to present the current state of knowledge concerning the harmfulness of chemical agents contained in wastewater and sewage sludge to human health and the surrounding environment. Wastewater and sewage sludge are composed of various chemical substances. These include volatile organic compounds (VOCs), persistent organic pollutants (POPs), heavy metals and pharmaceuticals. Inorganic gases, e.g. hydrogen sulphide, ammonia, are also emitted as a result of putrefaction processes occurring during wastewater treatment. Exposure to these diverse chemical agents poses a health risk due to their toxic, mutagenic and carcinogenic effects as well as their negative impact on endocrine functions. Because of the use of wastewater and sewage sludge in agriculture, they can be incorporated into the biological cycle, leading to the accumulation of pollutants in soils, vegetables and fruit as well as their transfer to surface water and groundwater. Not all chemical compounds are biodegradable in the environment, which contributes to their bioaccumulation and even biomagnification. The final recipient is man. While standards set for the content of specific pollutants in sewage sludge used for agricultural purposes are now stricter, they are not always complied with. Furthermore, new chemical compounds (e.g. pharmaceuticals) are identified in wastewater and their elimination with traditional wastewater treatment technologies is more difficult.

Keywords: chemical agents, wastewater, sewage sludge, health, environment, threat

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INTRODUCTION

According to research and data from the World Health Organisation (WHO), at least one sixth of the world's population does not have access to clean drinking water. The cause of this problem is water pollution due to human activities, such as industry, agriculture or generation of domestic sewage. Water pollution with chemicals may have a negative influence on both the environment and human health, which is why removal (disposal) of wastewater before the reuse of water is extremely important for general well-being (Act 2014, Alrumman et al. 2016, Tiwari et al. 2016).

In Poland, the current legislation provides precise definitions of wastewater and sewage, and specifies conditions to be met while discharging treated wastewater to water or land (Act 2017, Regulation 2014, 2019).

Wastewater constitutes a mixture of various water pollutants, including numerous liquids, solids, heavy metals, pesticides, radioactive compounds, pharmaceuticals, colloids and suspended solids as well as microorganisms. In the most general terms, we can define them as a mixture of various pollutants: physical, chemical and biological ones, coming from households, industrial activities and surface runoff, characterised by various degrees of pollution (Jurczyk et al. 2017, Sakson et al. 2017, 2018, Liang et al. 2019, Gworek et al. 2020).

While the removal of mechanical (visible) impurities is no longer a major problem, the elimination of invisible factors at wastewater treatment plants can be problematic. Apart from various biological contaminants that may be present in wastewater, chemical substances represent the largest group of invisible factors. Their source in municipal wastewater may be household pollutants, such as detergents or active ingredients of medicines and cosmetics. However, the vast majority of chemical pollutants originate from industrial plants. These include persistent organic pollutants (POPs), such as dioxins (PCDDs), furans (PCDFs), polycyclic aromatic hydrocarbons (PAHs) and heavy metals. Also, numerous chemicals from surface runoffs may be present in wastewater. Depending on the degree of pollution in the precipitation region, the chemical composition of rainwater runoff may vary (Mohd Udaiyappan et al. 2017, Sakson et al. 2017, 2018, Wontorska, Wąsowski 2018, Liang et al. 2019, Chmielewski et al. 2020, Gworek et al. 2020, Muhammad et al. 2021).

The problem of rational wastewater management in Poland still awaits solving, which is reflected in the annual reports of Statistics Poland (Polish abbreviation: GUS) relating to environmental protection. Analysis of the data confirms that the amount of generated wastewater is increasing each year; however, the effectiveness of wastewater treatment is not decreasing. The scale of sewage disposal and treatment in Poland in the selected years between 2014 and 2020 is illustrated by the data presented in Table 1 (GUS 2017, 2018, 2019, 2020).

Table 1

Treated and untreated wastewater discharged through sewage network (GUS 2017, 2018, 2019, 2020)

Specification	2014	2015	2016	2017	2018	2019	2020
	in cubic hectometres (hm^3)						
Municipal wastewater requiring treatment	1,238	1,258	1,290	1,317	1,330	1,343	1,344
Treated	1,236	1,254	1,289	1,316	1,329	1,337	1,344
Mechanically	0.4	0.4	0.4	0.4	0.4	0.3	0.3
Biologically	187.8	189.9	197.2	207.6	200.5	202.0	210.3
With increased removal of biogenic compounds	1,048	1,064	1,091	1,108	1,128	1,134	1,123
Untreated wastewater	1.5	4.1	1.2	1	1.6	6.7	10.4

The source: developed by the authors, based on data of GUS

During the treatment process of industrial and municipal wastewater, specific waste in the form of sewage sludge is generated that requires appropriate management due to its properties. The amount of dry mass in sewage sludge generated from industrial and municipal sewage treatment plants in the years 2019-2020 was 1,048.7 and 989.5 thousand tonnes, respectively, while the amount of sewage sludge generated at municipal sewage treatment plants was 574.6 and 568.8 thousand tonnes, respectively, and accounted for 55% and 58% of the total mass of the sludge generated in a particular year. Table 2 summarises the quantities of the sludge produced in the years 2013-2018 as compared to the population served by sewage treatment plants (GUS 2017, 2018, 2019).

According to the current legal regulations in Poland, municipal sewage sludge constitutes waste from sewage treatment plants, sludge from digesters and other installations used to treat municipal sewage, and other sewage with the composition similar to the one of municipal sewage (Act 2012).

Table 2

The amount of sewage sludge produced in the years 2013-2018 as compared to the population served by sewage treatment plants (GUS 2017, 2018, 2019)

Year	2013	2014	2015	2016	2017	2018
Produced sludge (thous. Mg/year)	526.7	519.2	533.3	540.3	556.0	568.0
Population of the cities and villages served by sewage treatment plants (thous.)	24,963	25,308	26,440	27,073	27,500	27,953

The source: developed by the authors, based on data of GUS

The specific properties of sewage sludge result mean that its thermal utilisation is not an indifferent process for the environment and – consequently - for health. This is due to the emissions of CO, SO_2, NO_x, dusts, PAHs, PCDDs/Fs as well as the risk of accumulated loads of heavy metals in the ash. On the other hand, management of sewage sludge, particularly in medium-size and small wastewater treatment plants, is still a significant problem in Poland because of the content of harmful chemicals and the necessity to handle them safely (Bolan et al. 2014, Grobelak et al. 2016).

As data presented by Statistics Poland (GUS 2020), the amount of produced sludge is about 1-3% of the volume of flowing wastewater; nevertheless it may pose a threat to health and the environment in the event of its improper management since it may contain heavy metals and pathogenic organisms. Table 3 presents treatment of sludge from municipal sewage treatment plants in the years 2013-2018 (GUS 2017, 2018, 2019).

Table 3

Management of sludge from municipal sewage treatment plants in the years 2013-2018 (GUS 2017, 2018, 2019)

Manner of management	2013	2014	2015	2016	2017	2018
	(thous. Mg d.m./year)					
Used in agriculture	105.4	107.2	107.5	116.0	108.5	118.3
Reclamation of areas	29.4	22.0	19.2	20.1	19.7	17.4
Use in the composting process	32.6	46.3	47.1	31.8	25.9	25.2
Subject to thermal recycling	72.9	84.2	79.3	101.1	106.2	111.5
Stored	31.4	31.5	40.5	20.7	15.3	10.6
Used in agriculture	105.4	107.2	107.5	116.0	108.5	118.3

The source: developed by the authors, based on data of GUS

SELECTED CHEMICAL AGENTS AND THE RISKS RELATED TO THEM

Odorous inorganic gases and volatile organic compounds

Decomposition of organic matter in wastewater and sewage sludge results in the emission of odorous substances (odours). These include odorous inorganic gases and volatile organic compounds (VOCs) that contain a wide range of chemicals. Odorous inorganic gases include hydrogen sulphide, ammonia, sulphur dioxide, n-methylenetetanamine, hydroxybenzene, cadaverine, putrescine, 3-methylindole, trimethylamine. VOCs include industrial solvents, toluene, benzene, esters, methane, trichloroethylene, chloroform, paraffin and ketones, hydraulic fluids, oil-based fuels, paints, inks and dry cleaners, aromatic compounds, aldehydes, ketones, carboxylic acids, esters, amines, aliphatic hydrocarbons, aromatic hydrocarbons, volatile organic sulphur compounds and organic aerobic compounds. The health and environmental effects of odorous inorganic gases and VOCs are widely described in the literature (Yang et al. 2019, Liang et al. 2020, Lomonaco et al. 2020, Chmielewski et al. 2020*a*, Słupek et al. 2020).

Hydrogen sulphide (H_2S) is a colourless gas, heavier than air, with the smell of rotten eggs, well-dissolving in the water, forming hydrogen sulphide water or, in higher concentrations, hydrogen sulphide acid. The smell is very strong and distinctly perceptible, even at very high dilution of 1/100,000 (1 cm^3 H_2S per 100 dm^3 air). It has a very toxic effect on aquatic organisms. In Poland, the threshold limit values (TLV) and short-term exposure limit (STEL) have been determined for hydrogen sulphide at 10 and 20 mg m^{-3}, respectively. Occupational poisoning with hydrogen sulphide is relatively common. The toxic effect of hydrogen sulphide is linked to blocking the activity of metal-containing enzymes in the prosthetic group. In the cells, hydrogen sulphide blocks the active iron of cytochrome oxidase, the final enzyme of the respiratory chain in mitochondria, and the activity of carbonyl anhydrase. The most sensitive tissues to hydrogen sulphide are mucous membranes and tissues with high oxygen demand (nervous tissue and cardiac muscle). The main target organs in acute hydrogen sulphide poisoning are the central nervous system and the lungs. Hydrogen sulphide in high concentrations (more than 4000 mg m^{-3}) causes death of animals within a few to several seconds. The respiratory system is affected – cyanosis, dyspnoea and death occur. After exposure to lower concentrations of hydrogen sulphide, conjunctivitis and painful corneal erosions appear immediately, the nose and throat are irritated and bronchitis occurs. Common complications include bronchopneumonia and pulmonary oedema. As a result of acute poisoning, a significant number of cases of neurological and neuropsychological lesions have been reported. Available data suggest that hydrogen sulphide does not have mutagenic, carcinogenic or teratogenic effects. However, recent studies confirm that it may play a potential role in pituitary cancer cells and is fully involved in the pathological process of tumours of the central nervous system (CNS), particularly gliomas and pituitary tumours (Mard et al. 2016, Cao et al. 2019, Ye et al. 2020, Wang et al. 2021, Peng et al. 2022).

Ammonia (NH_3) is a colourless gas with a characteristic, sharp, penetrating smell. Its smell for the human nose is perceptible at a concentration of 3.68 mg m^{-3}. The threshold limit values for ammonia are set at 35 mg m^{-3}. After exposure to ammonia concentrations lower or equal to the above mentioned ones, adverse effects on humans were not observed. Taking into account different personal sensitivity of humans and standard values accepted in the European Union, the threshold limit values were determined at a weighted average value for an 8-hour working day, i.e. 14 mg m^{-3} (19.74 ppm) and the value of short-term exposure limit at 28 mg m^{-3} (39.48 ppm). Mild irritation may already occur at concentrations of 50-100 mg m^{-3}, concentrations of 400-700 mg m^{-3} cause immediate irritation to the eyes as well as the upper respiratory tract. On the other hand, concentration exceeding 2000 mg m^{-3} constitute a lethal dose and death may occur very quickly due to lung damage. It irritates the mucous membranes of the eyes and the respiratory tract, resulting in decreased lung function. In the event of acute poisoning, bronchospasm and laryngeal oedema as well as bronchopneumonia may occur. Associated symptoms include burning throat, cough, salivation, lacrimation and headaches. At concentrations >1500 mg m^{-3}, apnoea, wheezing, chest pain and circulatory collapse may occur. However, contact with skin and eyes results in burns. The long-term exposure to ammonia in the atmosphere can contribute to the occurrence of chronic respiratory infections. The available literature shows that ammonia does not have teratogenic, mutagenic or carcinogenic effects (Rogalewicz, Bajdur 2014, Ubowska 2016, Rutkowski et al. 2019).

Aldehydes and ketones, thiols, carboxylic acids, esters, amines (e.g. crotonaldehyde, ethanedithiol, butyric acid, butyl acetate, triethylamine) are usually liquids with a characteristic pungent odour (e.g. sweat, rancid butter). They are already perceptible at low concentrations: µg m^{-3}. They irritate the respiratory tract, skin and eyes. The most common symptoms of exposure include coughing, sore throat, nausea, irritation of the mucous membranes of the eyes. Some of these substances (e.g. butyl acetate) may also become the cause of neurological symptoms, i.e. headaches and dizziness, drowsiness, loss of consciousness (Kilanowicz et al. 2017, Król, Dudziak 2018, Zwoździak, Piechocka 2018, Brzeźnicki, Bonczarowska 2021).

Persistent organic pollutants (POPs), heavy metals, pharmaceuticals

In addition to substances generated primarily in fermentation processes, wastewater also contains a number of undetectable chemical substances, including large quantities of biogenic compounds, heavy metals (e.g. Pb, Cd, Hg, Ni, Zn, Cu, Cr), organic micro-pollutants: PCDDs, PCDFs, polychlorinated biphenyls (PCB), PAHs, adsorbable organic halides (AOX) or extractable organic halides (EOX), pesticides (DDT), detergents (LAS) and pharmaceuticals, whose decomposition in the water often take years. Chemicals in wastewater can cause long-term health effects as most of them have mutagenic and carcinogenic effects. They may also impair the endocrine functions of the body (Grobelak et al. 2016, Liang et a. 2019, Chmielewski et al. 2020*b*, Gworek et al. 2020).

Polycyclic aromatic hydrocarbons (PAHs) constitute a widespread group of environmental carcinogens formed during incomplete combustion of organic substances. Polycyclic aromatic hydrocarbons, among which there are also toxic or carcinogenic compounds, are often found in rainwater and snowmelt. Afterwards, polluted water in the form of sewage enters the sewage system and further the surface waters. It is estimated that there are several hundred chemical compounds classified as PAHs in the environment. They are formed in the processes of incomplete combustion of organic matter.

They include benzo(a)pyrene, phenanthrene, and anthracene. After entering the human body, they bind with proteins, fats and the DNA, and contribute to the production of reactive oxygen species (ROS). This facilitates the emergence of numerous mutations and initiates the development of neoplasms. They also have cytotoxic effects, damaging the structures of cellular macromolecules and impairing their functioning. They cause changes in the immune system (immunotoxic effects, e.g. disruption of thymocyte maturation; decrease in serum immunoglobulin levels). PAHs may also cause harm to the foetus (Topal et al. 2014, Samburova et al. 2017, Chmielewski et al. 2020*c*, 2020*d*).

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzo-p-furans (PCDFs) belong to the group of stable, environmentally widespread, chlorinated polycyclic aromatic compounds with strong toxic properties. There are 75 congeners of PCDDs and 135 congeners of PCDFs. Unlike other chloro-organic substances, they have never been produced on an industrial scale and have never found a practical application. They were introduced into the environment as unintentional trace pollutants. They constitute by-products of chlorination reactions taking place in the following industries: chemical, pulp and paper, pharmaceutical, metallurgical, etc. as well as in the combustion of fuels, industrial, municipal and synthetic waste, and are emitted from municipal landfills. PCDDs and PCDFs may also be formed in chemical, thermal, photochemical and enzymatic reactions. PCDDs/PCDFs have been detected in emissions from all combustion processes, irrespective of the fuel, thus thermal processes are generally considered to be their main source in the environment, particularly waste incineration processes. However, the problem of PCDD/ PCDFs is inextricably connected with all kinds of waste management (Chmielewski et al. 2020*e*).

PCDDs/PCDFs have been classified as contaminants of high potential toxicological hazard due to their bioaccumulation in the food chain, the risk of exceeding the maximum intake levels in humans and their adverse health effects. Long-term exposure, even to low doses, may increase the risk of adverse health effects, such as disorders of the endocrine system, immune system, reproductive system, occurrence of neoplastic diseases or neuro-behavioural effects. The health effects of exposure to PCDD/Fs may manifest themselves after many years and also in the next generations. Effects on the developing foetus and infants are particularly dangerous. Toxic effects of exposure to PCDDs/PCDFs on humans and animals are based on the same mechanism, and are related to the activation of the intracellular aromatic hydrocarbon receptor (Ah receptor), which triggers a complex cascade of biochemical reactions and various toxic effects. PCDD/PCDFs also belong to the so-called endocrine disruptors. In exposed individuals, they may cause changes in thyroid hormone concentrations and disrupt the menstrual cycle. They have also teratogenic effects: they increase likelihood of miscarriages and birth defects in foetuses. People exposed to their effects have an increased risk of cancer. PCDD/PCDFs also cause a decrease in the body's immunity (Domingo 2014, Alawi et al. 2018, Kanan, Samara et al. 2018, Chmielewski et al. 2020*e*).

Heavy metals – the heavy metal group includes all metals with a density >5 g cm^{-3}. The occurrence and circulation of heavy metals in the environment is a natural phenomenon. The most important sources of heavy metals in the environment include bedrock, industrial emissions, transport emissions, municipal management and agriculture. The most significant anthropogenic sources of pollution of soils with heavy metals include: mining and metallurgy of non-ferrous metals, metallurgical industry, chemical industry, waste storage, application of contaminated mineral (mainly phosphatic) fertilisers in high doses, application of waste lime for deacidification of soils, plant protection agents, fertilisation with sludge as well as surface run-off from roads with heavy traffic. The threat from heavy metals results directly from their movement in the water-soil-plant-animal-human food chain and the ability to accumulate in the last link, i.e. the human body. The intensive development of industries as well as economic and social life contribute to a constant increase in concentrations of these metals in the environment. The excessive presence of heavy metals has a negative impact on all elements of the environment, and poses a threat to human health and sometimes even life (Chmielewski et al. 2020*f*).

The problem of pollution of the environment with heavy metals in Poland, despite technological changes in industrial plants limiting the emission of pollutants, remains one of the priority issues of public and environmental health.

Numerous studies confirm that heavy metals may have a negative impact on living organisms depending on their type, concentrations in the environment and forms in which they occur. Heavy metals are highly toxic to living organisms. Exposure to heavy metals may lead to a number of diseases, including disorders of the cardiovascular system, liver, kidneys, blood, and the nervous and skeletal systems. Many heavy metals are also recognised carcinogens. Heavy metals constitute identified toxic agents disrupting the normal functioning of the human body. In animal and human organisms, they may cause, in particular, changes in protein synthesis and disruptions in ATP synthesis, which can result in serious lesions, including neoplasms. The toxic effects of metals on humans and animals are very wide. The most toxic heavy metals include Pb, Hg and Cd. Metals easily accumulate in specific organs and a carcinogenic effect occurs when the level of the metal in a specific organism reaches or exceeds a threshold dose. Frequently, the most vulnerable organs are those involved in detoxification or elimination of the metal. Therefore, heavy metals mainly attack the liver and kidneys. Furthermore, metal accumulation is often found in the bones, brain and muscles. Metals can cause immediate acute poisoning or chronic conditions. Heavy metals belong to chemicals whose adverse reproductive and developmental toxicity have been assessed in numerous epidemiological studies (Trojanowska, Świetlik 2017, Piekut et al. 2018, Chmielewski et al. 2020*b*, 2020*c*, 2020*d*, 2020*f*, Walosik et al. 2021).

Pharmaceuticals are a group of biologically active chemical compounds that include antibiotics, non-steroidal anti-inflammatory drugs (NSAIDs), psychotropic drugs, β-blockers, hormones. Although their effects at therapeutic doses are well-defined, the effects of long-term exposure to a mixture of different pharmaceuticals at very low concentrations (ng l^{-1}; μg l^{-1}) are not known. In these quantities, as a contaminant, they can be present in sewage, surface water and also in drinking water. Studies on aquatic animals confirm that even such low concentrations of medications (hormones) can disrupt the endocrine metabolism of fish, leading to feminisation of males, reduced fertility, thus threatening survival of the population. Furthermore, in the face of antibiotic contamination of waters, multi-drug resistant bacterial strains may be isolated, resulting in the emergence of "incurable infections" or infections characterised by long-term treatment (Gworek et al. 2019, Praveena et al. 2019, Gworek et al. 2020).

SUMMARY

The negative impact of a sewage treatment plant on the environment, employees of the plant and residents of the surrounding area is widely discussed in the literature. The threat to human health often plays a significant role here.

For the functioning of the ecosystems, release of pollutants through the natural use of sewage sludge for fertilising purposes and discharge of sewage into surface waters constitute the cause of ecological threats. Bearing in mind environmental and health safety, understood as deliberate actions carried out to prevent unfavourable environmental changes for people, and thereby taking care of the needs and health of future generations, it is clear that the supply of adequate quality water to the population is one of the fundamental challenges. This issue should be placed at the forefront of environmental protection activities because it influences not only our daily life and health but also global environmental changes.

The above review of the literature confirms that wastewater contains many different chemical substances that are harmful to human health. Some of the organic compounds present in wastewater may have harmful effects on the environment and on human health. Health effects may result from both environmental and occupational exposure to these agents.

Due to the use of sewage sludge in agriculture, there is a risk that harmful chemical substances may penetrate into the basic elements of the environment: soils, waters and plants, including crops, and consequently into the human body.

It is necessary to monitor the presence and quantify potentially dangerous compounds in sewage and surface water where sewage is discharged.

It would be advisable to undertake educational activities within the scope of water quality addressed to a wide range of recipients, and aimed at changing their way of thinking and creating a sense of responsibility for the environment, including the issues related to sewage generation and its treatment, which may bring the expected effect, i.e. improvement in water quality.

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