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Heavy metals concentration in sewage treated at the collective wastewater treatment plant

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Abstract: This paper focuses on the analysis of the concentration of heavy metals in sewage treated at the municipal collective two-stage wastewater treatment plant (WWTP). The quality of raw sewage, mechanically treated sewage and totally treated sewage were tested. Heavy metals concentrations in sewage were in order Zn > Mn > Cu > Ni > Pb > Cr > Cd. A subsequent treatment stages gradually reduced metals concentrations, but the role of a mechanical treatment was not significant. The order of percentage reduction as a result of the total mechanical-biological treatment was Cr > Cd > Zn > Cu > Pb > Ni > Mn, which means that it was not consistent with the order of metals concentrations. It was found that totally treated sewage were characterised by greater variability in metal concentrations than raw sewage and mechanically treated sewage. Among the tested elements, the exception was Mn, with not only the lowest percentage reduction, but also with the most even concentrations level (low or average variability), while Cd and then, Cr, were characterised by the greatest differences in concentrations. Additionally, the following dependence was found: the greater WWTP hydraulic load, the greater heavy metals concentrations. Importantly, sewage met the legal requirements regarding to the permissible levels of heavy metals concentrations.

Keywords: heavy metals, mechanical-biological treatment, raw sewage, treated sewage, wastewater treatment plant

INTRODUCTION

Due to the carcinogenicity and toxicity, even in trace amounts, heavy metals constitute a group of the most dangerous and undesirable pollutants. Because they are not biodegradable, they can enter the environment, posing a direct threat for humans and other living organisms (Qasem, Mohammed and Lawal, 2021). When it comes to the presence of heavy metals in sewage, they get through to them as a result of production processes in many different industrial sectors; these includes metal, metallurgical and electroplating industries, production of artificial fertilisers, plant protection products and textiles, as well as paper mills and tanneries. Before discharging the sewage to the collective sewage system, industrial facilities should pre-treat industrial sewage using technological systems located on site (Koc-Jurczyk, 2013). The inflow of rainwater is a significant source of heavy metals in sewage, where zinc, copper, lead and cadmium are the most often detected in the highest concentrations (Sakson, Brzezinska and Zawilski, 2018). Research by Mikulski, Krzywonos and Borowiak (2021) showed that rainfall runoff from roofs with metal elements was characterised by the highest heavy metals concentrations, compared with rainwater samples collected, among others, from roofs without metal elements, from roofs with metal elements, but covered with vegetated soil, from streets and parking lots, or from stormwater settlement tanks outlets.

The use of classical methods for removing heavy metals from sewage and water, i.e. coagulation, chemical precipitation, solvent extraction, reverse osmosis, ion exchange, may be associated with some technological limitations and problems. Therefore, more effective techniques for metals removing are still being sought, such as the use of carbon nanomaterials (Mikulski, Krzywonos and Borowiak, 2021). Selection of a method for heavy metals removing depends on the pollution load, efficiency of the process, costs, as well as the degree of complexity of this process (Odumbe, Murunga and Ndiiri, 2023).

As a result of sewage treatment processes, heavy metals are accumulated in sewage sludge. Both chemical phosphorus removal processes based on precipitation with aluminium or iron salts, as well as the activity of activated sludge microorganisms participating in biological treatment processes, contributes to the removal of heavy metals from sewage through its accumulation in sludge (Krupicz and Masłoń, 2016). Although sewage sludge is a treatment by-product, it can be used in agriculture due to the high soil-forming and fertilising properties. Because the ecological risk posed by the presence of heavy metals in sewage sludge, the permissible content of Cd, Cu, Ni, Pb, Zn, Hg and Cr in sewage sludge that may be used for agricultural purposes or for land recultivation is formally limited (Tytła and Widziewicz-Rzońca, 2021). The possibility of sewage sludge reuse has focused the research of many scientists. For example, Kowalik *et al.* (2020) addressed the issue related to the mobility of heavy metals in sewage sludge. The tested by them Cu, Cr, Cd, Ni, Pb and Zn, were found mostly in non-mobile fractions, from which, metals not get into the soil. In turn, Szymański, Janowska and Jastrzębski (2011) proved that the content of heavy metals increases in thickened and dewatered sewage sludge compared to the metals concentration in raw sewage. Similarly, Tytła, Widziewicz and Zielewicz (2016), based on the conducted research found the highest metals concentration after sludge stabilisation, dewatering and hygienisation, and the dominance of immobile heavy metals fractions over mobile ones. At last, as Das and Poater (2021) show, using the partition coefficient *Kd* (this is the ratio of suspended solids to the dissolved solids in the aqueous phase), it is possible to determine, which heavy metals are more in sewage sludge and which in sewage liquid phase. According to the order of *Kd* coefficients calculated for different metals, where: Fe > Cd > Pb > Cr > Ni > Mn, it was concluded that among of them, the highest concentration in sewage liquid phase concerns Mn, while the highest content in sewage sludge is attributed to the Fe.

Based on the analysed literature, it was stated that many scientific studies concern the research on heavy metals in sewage sludge; this is due to the possibility of sewage sludge reusing, e.g., for agricultural purposes. However, within this paper, the authors were going to pay attention to the total heavy metals concentration in sewage undergoing treatment processes at the selected municipal collective wastewater treatment plant (WWTP). Through this analysis, the authors were going to complement the state of knowledge in this regard. For the needs of this paper, the other available research works related to the issue of heavy metals concentration in sewage were reviewed. After this, some selected research findings of the other authors were described and summarised in Table S1. Research on the changes in the quality of sewage during two-stages treatment has already been undertaken, however, with respect to oxygen pollutant indicators (biochemical oxygen demand – BOD_5, chemical oxygen demand – *COD*), total suspended solids or biogenic compounds (total nitrogen, total phosphorus) (Młyńska, Chmielowski and Młyński, 2017; Młyński *et al.*, 2020). Therefore, in this paper, a similar analysis related to the changes of metals concentration at the subsequent treatment stages was undertaken.

MATERIALS AND METHODS

RESEARCH OBJECT DESCRIPTION

The results of measurements of heavy metals concentration in sewage treated at the municipal collective wastewater treatment plant (WWTP) comes from the period 2017–2020. The analysed mechanical-biological WWTP is located in the southern Poland (49°39'30.6''N, 20°41'3.84''E) and serves the city area and neighbouring communes. The area of the agglomeration is covered by a 76 km-length combined sewage system and a separate sanitary sewage system, while the combined sewage system, constitutes only about a 13% of the total length of the sewer network, operated mainly as a gravity system. Between 2017 and 2020, the sanitary sewage system was expanded by a 30 km, reaching in 2020 a total length of 534 km. A population equivalent (p.e.) of residents using the sewage system in 2017 was 98,921 and 105,312 in 2020; a population equivalent of industry using sewage system was 60,305 in 2017 and 61,251 in 2020. While in 2017 about 11,000 of residents used septic tanks, in 2020, it was a two times less; domestic sewage treatment plants operate less than 500 residents.

The analysed WWTP (180,000 p.e.) receives domestic sewage and industrial sewage from small, local business entities. Maximum design hydraulic capacity of the WWTP is 33,000 $m^3 \cdot d^{-1}$. Raw sewage flowing into the WWTP, along with sewage delivered to the sewage decantation station, at first, are subjected to a mechanical treatment on two automatic bar screens, then, in two parallel grit chambers, and, in the final stage of mechanical treatment, in two radial primary settling tanks. The subsequent biological treatment takes place using activated sludge technology in two biological reactors (type MUCT), with separate anaerobic, hypoxic and aerobic zones. The clarified sewage outflow from the two radial secondary settling tanks located at the end of the WWTP technological line is discharged to the river (49°39'31.9''N, 20°40'45.8''E), which is a right tributary (a second-order river) of the Vistula River. Based on the data from the period 2017–2020, average water quality parameters of the receiving river, above (a) and below (b) sewage discharge point, were as follows: BOD_5(a) and BOD_5(b) < 3.0 mg $O_2 \cdot dm^{-3}$, *COD*(a) and *COD*(b) < 30.0 mg $O_2 \cdot dm^{-3}$, N_{tot}(a) = 3.80 mg$\cdot dm^{-3}$, N_{tot}(b) = 3.88 mg$\cdot dm^{-3}$, P_{tot}(a) = 0.075 mg$\cdot dm^{-3}$, P_{tot}(b) = 0.078 mg$\cdot dm^{-3}$, *TSS*(a) = 8.67 mg$\cdot dm^{-3}$, *TSS*(b) = 9.18 mg$\cdot dm^{-3}$, *EE*(a) and *EE*(b) < 5.0 mg$\cdot dm^{-3}$, where: BOD_5 = biochemical oxygen demand, *COD* = chemical oxygen demand, N_{tot} = total nitrogen, P_{tot} = total phosphorous, *TSS* = total suspended solids, *EE* = ether extract.

RESEARCH DATA HANDLING AND STATISTICAL ANALYSIS

Research material includes the results of measurements of total concentration for the seven selected heavy metals: zinc (Zn), lead (Pb), cadmium (Cd), copper (Cu), total chromium (Cr), nickel (Ni) and manganese (Mn) in sewage at the three treatment stages, i.e. in raw sewage flowing into the WWTP, in mechanically treated sewage and in totally treated sewage. These results were obtained from the WWTP operator and come from the period 2017–2020. Over this period, for each heavy metal and for each stage of treatment, about 90 sewage samples were tested for the total metal concentration. The concentrations of heavy metals were determined in the laboratory, using atomic absorption spectrometry with a graphite furnace according to the standard PN-EN ISO 15586:2005 (Polski Komitet Normalizacyjny, 2005) (lower concentrations) and by atomic absorption spectrometry with flame atomisation according to the standard PN-ISO 8288:2002 (Polski Komitet Normalizacyjny, 2002) (higher concentrations). Chromium, lead, copper and nickel were determined according to the PN-EN ISO 15586:2005 and the PN-ISO 8288:2002 (0.005÷0.02 mg·dm^{-3}), cadmium according to the PN-EN ISO 15586:2005 and the PN-ISO 8288:2002 (0.0005÷0.002 mg·dm^{-3}), zinc according to the PN-ISO 8288:2002 (0.2÷1.0 mg·dm^{-3}), and manganese, according to the PN-EN ISO 15586:2005 (0.2÷1.0 mg·dm^{-3}) and additionally according to the standard PN-92/C-04570.01 (Polski Komitet Normalizacyjny, 1992), describing the atomic absorption spectrometry method without pre-concentration (0.005÷0.02 mg·dm^{-3}).

All the calculations and graphs were elaborated using MS Excel program. As part of the initial analysis, based on the data shared of average hourly sewage inflows in particular days of the period 2017–2020, daily sewage WWTP hydraulic loads were examined. For this purpose, some parameters, including minimum (min.), maximum (max.), average (avg.), standard deviation (*SD*), coefficient of variation (*CV*) and the number of exceedances (N_e) of the WWTP hydraulic capacity (33,000 m^3·d^{-1}), were determined. For each metal concentration at the subsequent treatment stages, basic descriptive statistics for the period 2017–2020 were determined; these includes min., max., avg., *SD* and *CV*. Additionally, separately for each year of the multi-year period, the average yearly heavy metals concentrations at each stage of treatment were determined. For each heavy metal, the average and standard deviation of the percentage reduction (η) for totally treated sewage, were determined using the Equation (1) (Szczerbińska *et al.*, 2018).

$$\eta = [(C_{\text{raw}} - C_{\text{ttreated}})/C_{\text{raw}})] \cdot 100\% \quad (1)$$

where: η = percentage reduction of heavy metals for totally treated sewage (i.e. sewage treatment efficiency) (%), C_{raw} = heavy metal concentration in raw sewage (mg·dm^{-3}), C_{ttreated} = heavy metal concentration in totally treated sewage (mg·dm^{-3}).

Variability of the WWTP hydraulic load and the variability of heavy metals concentration were examined using *CV*. The values of *CV* were interpreted as follows: below 0.25 = low variability, between 0.25 and 0.45 = average variability, between 0.45 and 1.00 = strong variability, above 1.00 = very strong variability. Heavy metals concentrations in sewage were compared with the permissible level in treated sewage, as the Polish Regulation (Rozporządzenie, 2019) determines: Zn = 2.0 mg·dm^{-3}; Pb, Cu, Cr, Ni = 0.5 mg·dm^{-3}; Cd = 0.2 mg·dm^{-3}; Mn = not specified.

RESULTS

HYDRAULIC LOAD OF THE WASTEWATER TREATMENT PLANT

At the beginning of the research, hydraulic load of the wastewater treatment plant (WWTP) was examined. This is because hydraulic load is one of the factors influencing the effectiveness of pollutants removal. For the multi-year period 2017–2020, the average sewage inflow was 23,600 m^3·d^{-1} (Fig. 1), which means, it was smaller than the WWTP hydraulic capacity of 33,000 m^3·d^{-1} by about 30%. As it can be seen in the Figure 1, in the period 2017–2020, exceedances of the WWTP hydraulic capacity were noted. Among the investigated four years, 2019 and 2020 turned out to be the years with the greatest average sewage inflow (around 24,000–25,000 m^3·d^{-1}), the greatest standard deviation of sewage inflows (over 7,000 m^3·d^{-1}), the greatest variability of sewage inflow (*CV* about 0.30, i.e. average variability) and the greatest number of the WWTP hydraulic capacity exceedances (over 40) (Tab. 1). In turn, 2018 is considered as the year with the most stable sewage inflow conditions, what is evidenced by the values of average sewage inflow (avg. = 21,289 m^3·d^{-1}), standard deviation of sewage inflow (*SD* = 4,722 m^3·d^{-1}), coefficient of variation (*CV* = 0.22, i.e. low variability of sewage inflows) and only 10 records for exceedances of the WWTP hydraulic capacity (Tab. 1). As a direction for future research, an additional precise monitoring of sewage flows and weather conditions is taken into consideration in order to divide into the dry and rainy periods, which would provide a better insight into the presented issue. At now, these results were presented in order to show only some general trends for the WWTP hydraulic load and the heavy metals concentration.

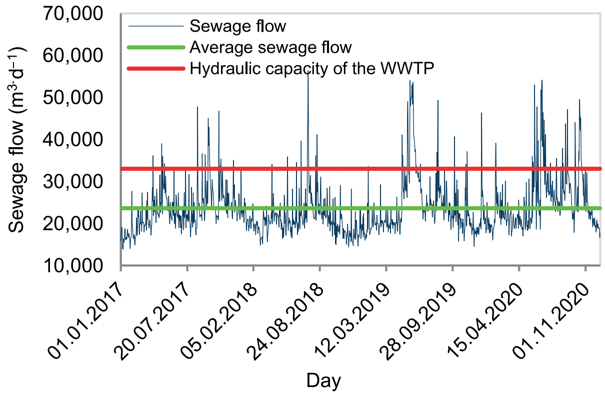


Fig. 1. Hydraulic load of the wastewater treatment plant (WWTP) in the period 2017–2020; source: own study

Table 1. Descriptive statistics for hydraulic load of the wastewater treatment plant (WWTP) in particular years of the period 2017–2020 along with the number of exceedances of hydraulic capacity

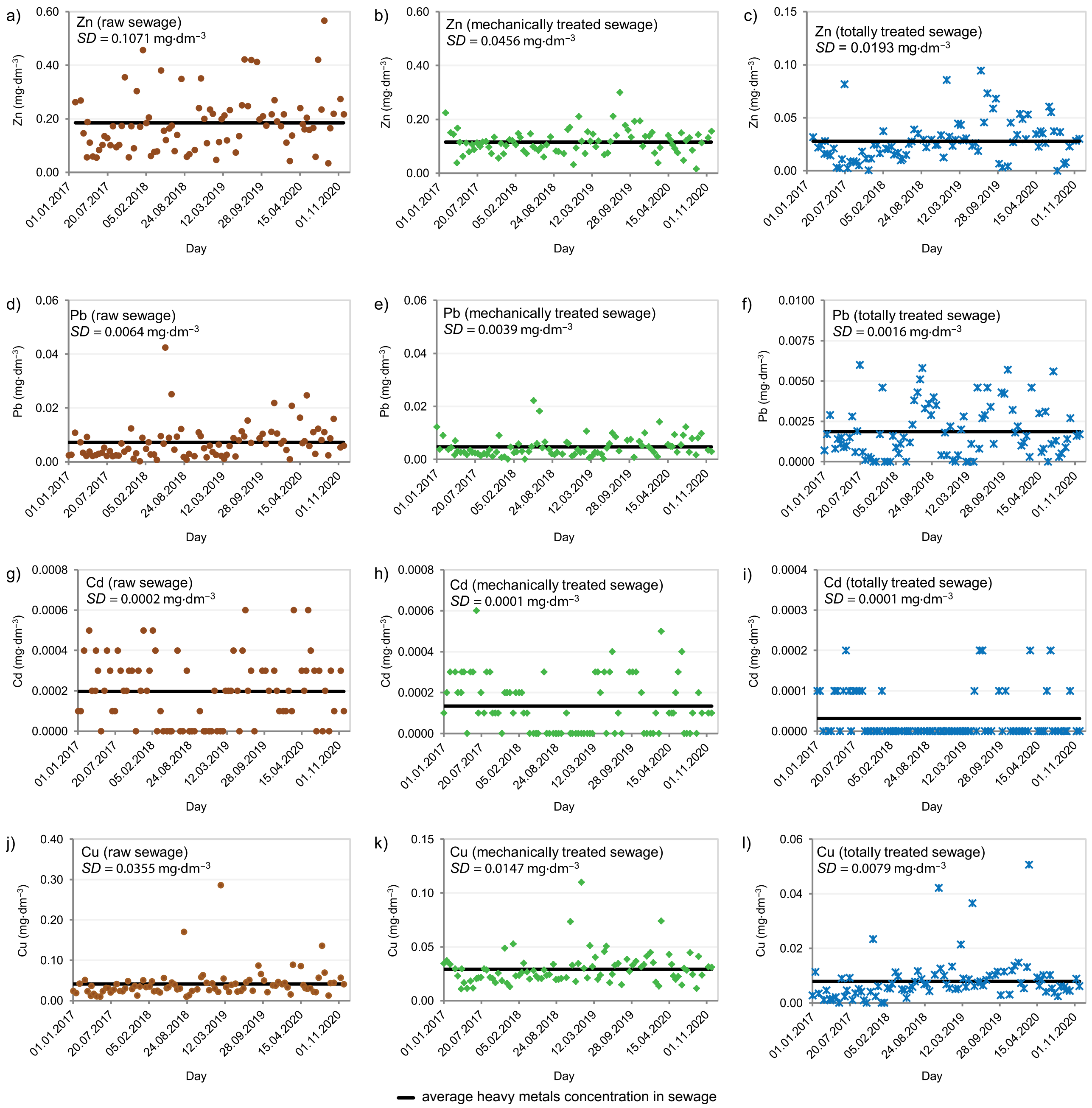
Parameter	Unit	Year			
		2017	2018	2019	2020
Min.	m^3·d^{-1}	13,920	14,384	14,485	16,028
Max.		47,760	56,383	54,048	54,123
Avg.		23,327	21,289	24,421	25,495
SD		5,184	4,722	7,437	7,130
CV	–	0.22	0.22	0.30	0.28
N_e		21	10	41	43

Explanations: min. = minimum, max. = maximum, avg. = average, *SD* = standard deviation, *CV* = coefficient of variation, N_e = number of exceedances of the wastewater treatment plant hydraulic capacity.
Source: own study.

HEAVY METALS CONCENTRATION IN SEWAGE AT THE SUBSEQUENT TREATMENT STAGES

Out of the seven heavy metals analysed, in raw sewage flowing into the WWTP in the period 2017–2020, the greatest amounts referred to the Zn (avg. = 0.185 mg·dm^{-3}, *SD* = 0.107 mg·dm^{-3}) and Mn (avg. = 0.089 mg·dm^{-3}, *SD* = 0.025 mg·dm^{-3}), while the least amounts, were noted for Cd (avg. = 0.0002 mg·dm^{-3}, *SD* = 0.0002 mg·dm^{-3}) (Fig. 2, Tab. 2). The values of coefficients of variation (*CV*) (Tab. 2) indicate that out of the all treatment stages, totally treated sewage were characterised by the greatest variability in heavy metals concentrations (strong or very strong variability). Despite a raw sewage and sewage after mechanical treatment had a lower *CV* values, they still indicated strong or very strong variability. The exception was Mn, for which, after mechanical treatment, the *CV* value indicated low variability, while in raw sewage and in totally treated sewage – average variability. Total treatment processes provided the greatest reduction for Cr (avg. = 87.5%, *SD* = 16.9%) and the lowest reduction for Mn (avg. = 41.5%, *SD* = 18.3%). For the other metals analysed, the percentage reduction was in the range between 64.8% (Ni) and 82.4% (Cd) (Tab. 3). Although the Figure 2 shows that over the subsequent treatment stages, heavy metals concentrations in general gradually decreased, the percentage reduction was not elaborated for mechanically treated sewage. This is because in many samples after mechanical treatment, heavy metals concentration was higher than in raw sewage. Depending on the metal, this affected from about 15% to about 40% of the tested samples (to the greatest extend, for Cd). This may indicate, for example, the release of metals from sewage sludge into the liquid phase of sewage.

In graphs presented in the Figure 3, again one can primarily observe a gradual decrease in heavy metals concentration in



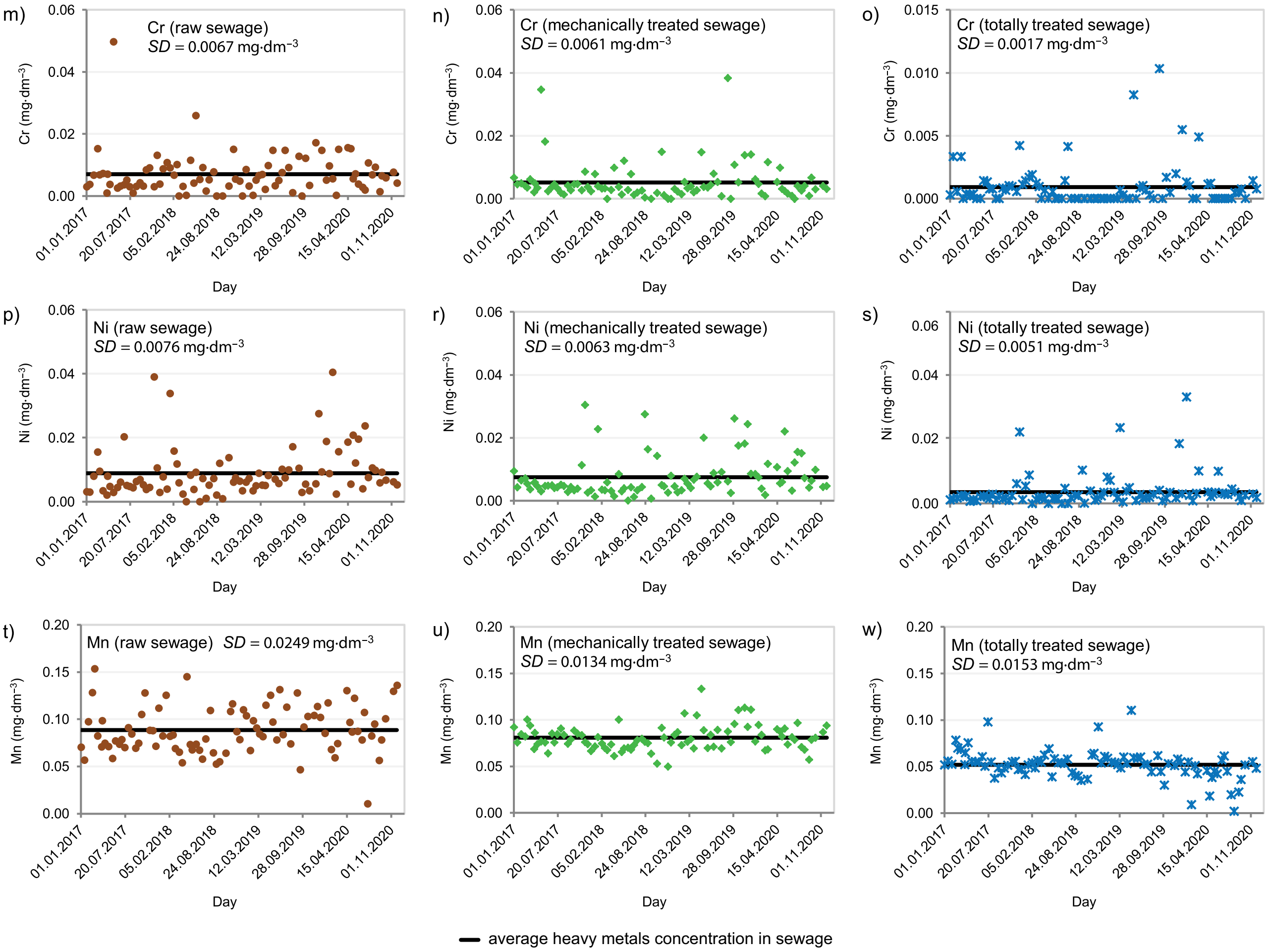


Fig. 2. Heavy metals concentrations in sewage at the subsequent treatment stages in the period 2017–2020 along with average concentrations and standard deviations (*SD*); source: own study

Table 2. Descriptive statistics for heavy metals concentrations in sewage at the subsequent treatment stages in the period 2017–2020

Parameter	Unit	Element						
		Zn	Pb	Cd	Cu	Cr	Ni	Mn
Raw sewage								
Min.	mg·dm^{-3}	0.034	0.000	0.000	0.001	0.000	0.000	0.011
Max.		0.567	0.043	0.0006	0.286	0.050	0.041	0.154
Avg.		0.185	0.007	0.0002	0.041	0.007	0.009	0.089
SD		0.107	0.006	0.0002	0.036	0.007	0.008	0.025
CV	–	0.58	0.89	0.84	0.86	0.95	0.86	0.28
Mechanically treated sewage								
Min.	mg·dm^{-3}	0.016	0.000	0.000	0.011	0.000	0.000	0.050
Max.		0.300	0.022	0.0006	0.110	0.038	0.030	0.134
Avg.		0.116	0.005	0.0001	0.029	0.005	0.008	0.081
SD		0.046	0.004	0.0001	0.015	0.006	0.006	0.013
CV	–	0.39	0.82	1.02	0.50	1.18	0.84	0.17
Totally treated sewage								
Min.	mg·dm^{-3}	0.000	0.000	0.000	0.000	0.000	0.000	0.002
Max.		0.094	0.006	0.0002	0.051	0.010	0.033	0.110
Avg.		0.028	0.002	0.0000	0.008	0.001	0.004	0.052
SD		0.019	0.002	0.0001	0.008	0.002	0.005	0.015
CV	–	0.70	0.87	1.85	1.00	1.77	1.34	0.30

Explanations as in Tab. 1. Source: own study.

Table 3. Percentage reduction of heavy metals concentrations in totally treated sewage in the period 2017–2020

Parameter	Unit	Element						
		Zn	Pb	Cd	Cu	Cr	Ni	Mn
η_{avg}	%	81.0	75.4	82.4	76.3	87.5	64.8	41.5
η_{SD}		16.1	20.6	28.5	19.2	16.9	20.4	18.3

Explanations: η_{avg} = average percentage reduction, η_{SD} = standard deviation of percentage reduction.
Source: own study.

sewage at the subsequent treatment stages. For all tested heavy metals, their concertation before and after treatment processes was lower than the permissible values, as the Regulation (Rozporządzenie, 2019) determines, i.e. 2.0 mg·dm^{-3} (Zn), 0.5 mg·dm^{-3} (Pb, Cu, Cr, Ni) and 0.2 mg·dm^{-3} (Cd); only for Mn, a limited concentration in sewage is not specified formally. As it can be seen in the Figure 3, particular years of the period 2017–2020 differed of the average concentration of heavy metals. Except of the Mn, these differences concerned in the slightest extent to the totally treated sewage. In most cases, 2019 was the year with the highest average heavy metals concentration, while the lowest average heavy metals concentration, can be attributed to the 2017 or 2018. The greatest differences in heavy metal concentrations between particular years may be attributed to the Cd (Fig. 3c) and in the second order, to the Cr (Fig. 3e), while the Mn concentrations (Fig. 3g), remained the most uniform over the four-year period.

Linking to the results of heavy metals concentration with the measurements of sewage inflows in particular years of the period 2017–2020 (Tab. 1), some dependence can be stated: the greater WWTP hydraulic load, the greater heavy metal concentration. Increased hydraulic loading may cause an increase in the concentration of heavy metals in sewage due to the shortening of the retention time in biological reactors, excessive increase in flow velocity and loading of settling tanks, limiting the efficiency of sedimentation and chemical and biological processes. It suggests the need for empirical analysis of the correlation between flow and metals concentration, optimisation of sedimentation processes and implementation of flow management strategies to improve treatment efficiency.

As it was presented above, sewage treatment ensured the required level of heavy metals removal, while meeting the requirements of Regulation (Rozporządzenie, 2019). As it was recorded, metals concentrations in sewage were in order, from the highest, Zn > Mn > Cu > Ni > Pb > Cr > Cd. Similar relationships can be found, e.g., in the paper of Chipasa (2003), where the metals concentrations were in order Zn > Cu > Pb > Cd. In the study by Mansourri and Madani (2016), these dependences were in order Ni > Cu > Pb > Zn > Cr, in the study by Olujimi *et al.* (2012), it was Zn > As > Cd > Hg and in the paper of Kulbat *et al.* (2003), it was Zn > Cu > Pb > Cr > Ag > Ni > Cd. When it comes to the obtained percentage reduction as a result of the total mechanical-biological treatment, this is in order, from the highest, Cr > Cd > Zn > Cu > Pb > Ni > Mn. While, for example, Chipasa (2003) found in his research that the higher metal concentration, the higher percentage reduction, the

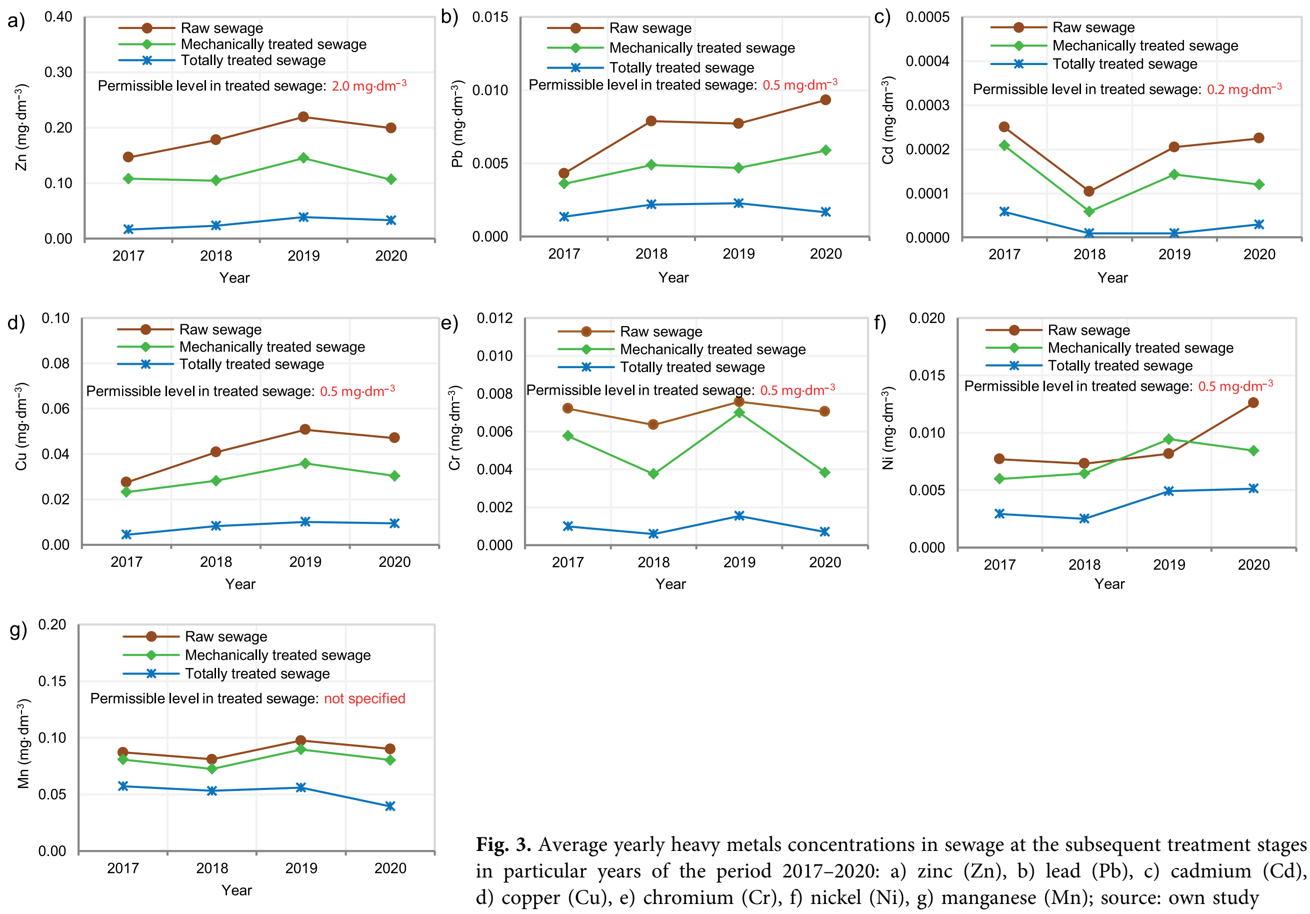


Fig. 3. Average yearly heavy metals concentrations in sewage at the subsequent treatment stages in particular years of the period 2017–2020: a) zinc (Zn), b) lead (Pb), c) cadmium (Cd), d) copper (Cu), e) chromium (Cr), f) nickel (Ni), g) manganese (Mn); source: own study

research results presented in our paper, are not consistent with these reports: the order of the achieved percentage reduction, from the highest, was not the same like the order of metals concentrations. Another research by Olujimi *et al.* (2012) showed that after mechanical treatment, significant heavy metals reduction concentration can be achieved. Kulbat *et al.* (2003), however, state the essential role of biological treatment and a small share of mechanical processes in metals removal (mechanical processes ensured a reduction of at most 20%, while mechanical-biological treatment resulted in a total reduction of 80–90%). Similarly like Kulbat *et al.* (2003), in this paper, a dominant role of biological treatment in heavy metals removal was observed.

CONCLUSIONS

The knowledge in the field of heavy metals concentrations in sewage treated at collective wastewater treatment plants (WWTP) is important from the point of view the correctness of treatment processes and thus, the protection of natural receivers. In this study, in order to supplement the state of knowledge in this regard, seven chemical elements were analysed; these included Zn, Pb, Cd, Cu, Cr, Ni and Mn. The concentrations of metals in sewage at the end of technological line of the tested municipal collective two-stage WWTP met the requirements regarding the permissible levels, as determined by the Polish Regulation (Rozporządzenie, 2019). Concentrations of the tested metals were in order Zn > Mn > Cu > Ni > Pb > Cr > Cd. Comparing the average concentration of Zn and Cd, it was observed that the Zn concentration was about nine hundred times higher than the Cd concentration. Although the subsequent treatment stages resulted in a gradual metals reduction, the role of a mechanical treatment is not significant. The greatest reduction as a result of the total mechanical-biological treatment processes was attributed to the Cr (avg. = 87.5%), while the lowest reduction, to the Mn (avg. = 41.5%). The obtained order of the percentage reduction Cr > Cd > Zn > Cu > Pb > Ni > Mn is not consistent with the order of the average metals concentrations as indicated above. It was observed that totally treated sewage were characterised by greater variability in metal concentrations than raw sewage or mechanically treated sewage; however, for all treatment stages, variability was still strong or very strong. Among the tested elements, the exception was Mn with a low or average variability. Over the years, Cd and then, Cr, were characterised by the greatest differences in concentrations, while the Mn concentrations, remained the most uniform over the four-year period. At last, linking the results of the WWTP hydraulic load with the heavy metals concentrations, their mutual proportionality was found, i.e. the greater hydraulic load, the greater heavy metals concentrations. Because it may be a consequence of shortening of the retention time in biological reactors or the excessive increase in flow velocity and loading of settling tanks, there is a suggestion for optimisation of sedimentation processes and implementation of flow management strategies to improve treatment efficiency.

SUPPLEMENTARY MATERIAL

Supplementary material to this article can be found online at: https://www.jwld.pl/files/Supplementary_material_66_Mlynska.pdf

CONFLICT OF INTERESTS

All authors declare that they have no conflict of interests.

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