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# APLICATION OF HEAVY METALS AND NUTRIENTS INTO NATURAL ENVIRONMENT WITH SEWAGE SLUDGE

## METALE CIĘŻKIE I SKŁADNIKI POKARMOWE WPROWADZANE DO ŚRODOWISKA PRZYRODNICZEGO Z OSADAMI ŚCIEKOWYMI

**Abstract:** The paper presents results of examinations of sludge from dairy sewage treatment plants in north-eastern Poland and results of examinations of compost obtained from research facilities applying low-expenditure methods of treatment of dairy sewage sludge with added structural materials. Chemical substances have been analyzed and their amount estimated, together with diary sewage sludge entering the environment. Admissible amounts of heavy metals contained in sewage sludge according to Polish and international legal regulations have been referred to.

Very low harmfulness of diary sludge has been stated. It has been demonstrated that the process of composting and vermicomposting has changed the structure of the sludge and additionally enriched it with fertilizer values (N, P, K content) and soil formation values.

**Keywords:** vermiculture, sewage sludge, low cost methods, compost, vermicompost, Dairy Industries

Sludge is the source of mineral and organic pollution. In recent years much attention has been paid to examining organic pollution. In sewage sludge 516 compounds have been identified and classified into 15 classes. Among them are *polycyclic aromatic hydrocarbons* (PAHs), *adsorbable organic halogens* (AOX), *polychlorinated biphenyls* (PCBs), and others. Besides these compounds, in sewage sludge can be found toxic trace elements which hamper sludge utilization. Sludge is also a source of organic matter, as well as macro- and microelements of agricultural significance, hence it can be applied in agriculture [1].

In the north-eastern part of Poland, sludge is more and more widely managed and exposed to low-expenditure treatment methods. Such activities have been influenced by the following factors: agricultural and industrial specificity of the region, sludge

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characteristics, biomass in form of straw, sawdust and chips present in the Green Lungs of Poland Region, soils of low class, and bigger amount of small and medium sewage treatment plants [2].

At present, in Podlasie province there are 113 municipal and 30 industrial sewage treatment plants of throughtput of several to several thousand cubic meters per day [3]. In the 1990's a significant progress was observed in the field of protection of water environment of the region. Many new facilities were constructed, capable of intense sewage treatment. Most serious negligence concerns industrial sewage treatment, in particular dairy workshops. Most new and modernized sewage treatment plants marginalized the problem of sewage sludge treatment by applying natural or mechanical appliances for sludge dewatering. Because of physical and chemical composition of sewage sludge from dairy industry, it should be considered as a raw material rather than waste. The fundamental criterion enabling sludge return to the natural environment is its sanitary condition. The author's research shows that dairy industry sludge basically does not contain pathogenic organisms, including bacteria, fungi, protozoa, parasites and their fertilized eggs, which would prevent their use for non-industrial purposes. Moreover, the sludge is characterized by very low content of heavy metals and other specific chemical compounds (very often present in municipal sewage sludge) as well as high fertilizing values; it is the source of carbon as well as nitrogen and phosphorus. At the same time, the waste is very often considerably liquefied, which makes its thorough distribution on the ground difficult. Safe application of the sludge in the environment without prior treatment becomes more and more limited because of legal regulations which limit, and in the nearest future will prevent, direct fertilizing application of untreated sewage sludge. On the other hand, soil structure of Poland and its north-eastern regions is dominated by light soils which are characterized by low content of nutrients, poorly developed sorptive complex, and an inconsiderable amount of humus. Entry to soils of fertilizer in form of treated sewage sludge from dairy industry and composts obtained with participation of other components should be considered as a factor which counteracts soil degradation and enhances soil fertility [2, 4].

## Material and methods

The article presents results of examinations of dairy sewage sludge in north-eastern Poland and results of examination of composts obtained from research facilities applying low-expenditure methods of treatment of dairy sewage sludge with added structural material.

The purpose of the examinations was to analyze and estimate chosen chemical pollutants entering the environment together with stabilized sewage sludge from dairy sewage treatment plants and treated by means of low-expenditure methods. The general concept of the examinations assumed comparing obtained results of examinations of the sludge with added components from chosen facilities treating sludge with the use of California earthworm and in the process of windrow composting. Obtained results included examinations carried out in dairy sewage treatment plants and in research facilities located in Rudka forest inspectorate and in a sewage treatment plant in Zambrow.

Examinations were carried out in research facility in technological scale in Rudka forest inspectorate in Korycin tree nursery where sewage sludge was composted in windrow composting technology with throwing of compost by means of a compactor. Basic structural materials used in the process of composting in the facility were sawdust, straw and chips. Another research facility in technological scale is located in a sewage treatment plant in Zambrow where sewage sludge is treated with the use of vermiculture. Beds with vermiculture were fed with the same dairy sewage sludge with liquefication of approx. 80 %, once, in the amount of 1.5 $m^3$ of liquefied sludge per bed of 4 $m^2$. Before being fed, the bed was embanked with the so-called lair of California earthworms (*Eisenia fetida*) coming from the sewage treatment plant in Zambrow. Earthworms consume sludge and excrete coprolites, ie vermicompost. Both facilities used stabilized sewage sludge from dairy sewage treatment plant of S.M. MLEKOVITA in Wysokie Mazowieckie [1, 5].

The analysis of obtained material included the following examinations: heavy metal content, fertilizing values, liquefication, dry mass, and reaction. The examinations were carried out in the Department of Technology in Engineering and Environmental Protection laboratory according to valid norms.

Sludge samples were treated with mineralization in HACH mineralizator with the use of sulphuric acid and hydrogen peroxide in mixture of nitric and hydrochloric acid in ratio 1:3. For further analysis mineralizats were filtered through MN 616 G paper filter.

Determination of cadmium, nickel and total chromium content was done in samples of mineralizats with the use of *atomic absorption spectrometr* Perkin-Elmer 4100 ZL with transversely heated graphite cuvette and Zeeman-effect background correction.

Determination of mercury content was done in samples of mineralizats by means of cold steam technique with the use of atomic absorption spectrometr Perkin-Elmer 4100 ZL equipped in add-on device FIAS-200.

Determination of zinc, lead and copper content was done in samples of mineralizats by means of flame atomization, and potassium content was done by means of *atomic emission spectroscopy* (AES) with the use of atomic emission spectrometr Varian SpectrAA 20 Plus.

Fig. 1. Flow diagram of the research installation

## Results and discussion

In the last decade, the amount of sewage sludge generated by the analyzed dairy sewage treatment plants increased considerably, and presently it amounts to approx. 3450 Mg of dry mass per year. In comparison, all municipal sewage treatment plants in the province produced 17409 Mg of sludge dry mass, out of which 6865 Mg of dry mass were applied in agriculture or for remediation. In 2009, global amount of sludge from industrial sewage treatment and pretreatment plants amounted to 7360 Mg, out of which 3550 Mg of dry mass were applied in agriculture or for remediation [3]. It implies that in the province approx. 40 % of municipal sludge and nearly 50 % of sewage sludge was environmentally applied. Thus, dairy sludge constitutes approx. 47 % of industrial sludge in the province and is environmentally used in almost 100 %. There was an increase of approx. 150 % in the amount of dairy sludge comparing with the amount in the mid 1990's.

Dairy sewage sludge is characterized by much lower heavy metal content than municipal sludge. Table 1 presents average amounts of heavy metals in stabilized sewage sludge from diary workshops in the region in the last 10 years, and composts obtained from sludge from the largest facility in Wysokie Mazowieckie.

Table 1

Heavy metals content in sewage sludge from dairy wastewater treatment plants (average values 2000–2009) and compost from Rudka Forestry research installation and Zambrow instalation

| Localization | The quantity of heavy metals [mg $\cdot$ kg$^{-1}$ d.m.] | | | | | | |
|---|---|---|---|---|---|---|---|
| | Pb | Zn | Cu | Cd | Ni | Cr | Hg |
| Sejny | 10.0 | 240 | 26 | 0.80 | 1.9 | 2.1 | 0.17 |
| Bielsk Podlaski | 5.0 | 348 | 24 | 0.42 | 12.1 | 13.1 | 0.11 |
| Zambrow | 8.1 | 230 | 28 | 0.92 | 9.1 | 9.6 | 0.23 |
| Wysokie Mazowieckie | 12.0 | 366 | 45 | 0.52 | 5.1 | 18.4 | 0.20 |
| Grajewo | 19.9 | 327 | 22 | 0.45 | 7.4 | 17.6 | 0.19 |
| Monki | 4.2 | 197 | 31 | 0.11 | 6.8 | 11.5 | 0.21 |
| Kolno | 12.6 | 139 | 17 | 0.15 | 8.0 | 6.9 | 0.06 |
| Piatnica | 7.1 | 410 | 15 | 0.26 | 7.9 | 14.8 | 0.10 |
| Rudka (compost) | 7.8 | 149 | 23 | 0.48 | 4.9 | 6.3 | 0.15 |
| Zambrow (vermicompost) | 6.2 | 195 | 22 | 0.34 | 3.8 | 7.9 | 0.14 |

Source: own data processing [1, 6].

Table 2 presents basic fertilizing values of sewage sludge from analyzed dairy sewage treatment plants.

All dairy sludge are characterized by high fertilizing value. Also characteristic is very high content of nitrogen and phosphorus compounds as well as organic substance. Most facilities do not apply the process of hygienization because of very good sanitary condition of the sludge. In order to obtain proper parameters of biomass exposed to the process of composting, according to assumed methodology and technology, dewatered

Table 2

Basic fertilizing values of sewage sludge from dairy wastewater treatment plants – average values 2000–2009

| Localization | Chosen fertilizing parameters | | | | | | |
|---|---|---|---|---|---|---|---|
| | Total N | Ammonia N | Total P | Magnesium | Calcium | Organic substance | pH |
| | [g · kg$^{-1}$ d.m.] | | | | | [%] | |
| Sejny | 69.2 | 2.8 | 69.1 | 5.2 | 17.5 | 61.2 | 7.32 |
| Bielsk Podlaski | 21.6 | 2.6 | 21.6 | 6.8 | 61.9 | 74.2 | 7.67 |
| Zambrow | 93.5 | 0.5 | 48.8 | 5.7 | 41.3 | 74.0 | 7.13 |
| Wysokie Mazowieckie | 92.6 | 2.9 | 22.5 | 3.9 | 28.2 | 82.9 | 12.71 |
| Grajewo | 34.9 | 11.5 | 10.6 | 1.0 | 24.9 | 67.1 | 7.19 |
| Monki | 62.5 | 6.4 | 64.0 | 2.1 | 18.0 | 64.3 | 6.64 |
| Kolno | 71.1 | 1.8 | 71.0 | 5.8 | 49.0 | 34.1 | 7.21 |
| Piatnica | 74.3 | 38.0 | 7.6 | 6.9 | 12.4 | 82.0 | 7.33 |

Source: own data processing.

dairy sewage sludge have been mixed with various structural materials. Table 3 presents heavy metal contents in materials mixed with sludge. Amounts of individual metals are very low.

Table 3

Heavy metals contents in origin material

| Substrate | Pb | Hg | Cu | Cd | Ni | Zn | Cr |
|---|---|---|---|---|---|---|---|
| | [mg · kg$^{-1}$ d.m.] | | | | | | |
| Straw | 9.0 | 0.018 | 10.0 | 0.18 | 4.5 | 20.0 | 4.0 |
| Saw dust | 10.0 | 0.06 | 4.8 | 0.20 | 5.0 | 12.5 | 2.3 |
| Wooden chip | 25.0 | 0.14 | 18.0 | 0.80 | 6.0 | 118 | 12.0 |

Source: [1].

Examinations of applied structural materials for biogenic elements and macroelements (Table 4) show, first of all, very high carbon content: 35 % to 44.5 %. Thus, it is a material which is supposed to regulate proportion C:N in compost mass. Content of the remaining components is less important regarding their content in dairy sludge.

Table 4

Origin material – fertilizing values

| Parameter | Unit | Substrate | | |
|---|---|---|---|---|
| | | straw | saw dust | wooden chip |
| Ca | [g · kg$^{-1}$ d.m.] | 6.6 | 6.7 | 2.9 |
| | [% CaO d.m.] | 0.92 | 0.94 | 0.4 |

Table 4 contd.

| Parameter | Unit | Substrate | | |
|---|---|---|---|---|
| | | straw | saw dust | wooden chip |
| Mg | [g · kg$^{-1}$ d.m.]<br>[% MgO d.m.] | 0.6<br>0.10 | 0.63<br>0.10 | 0.55<br>0.09 |
| Total N | [g · kg$^{-1}$ d.m.]<br>[% d.m.] | 7.14<br>0.71 | 4.28<br>0.43 | 12.3<br>1.23 |
| Ammonia N | [g · kg$^{-1}$ d.m.]<br>[% d.m.] | 0.61<br>0.061 | 1.30<br>0.13 | 0.80<br>0.08 |
| Total P | [g · kg$^{-1}$ d.m.]<br>% $P_2O_5$ d.m. | 5.36<br>2.46 | 1.85<br>0.85 | 1.43<br>0.65 |
| K | [g · kg$^{-1}$ d.m.] | 5.2 | 0.5 | 3.9 |
| TOC* | [g · kg$^{-1}$ d.m.]<br>[% d.m.] | 375.9<br>37.6 | 444.6<br>44.5 | 350.4<br>35.0 |

* Total Organic Carbon.
Source: [1]

Tables 5 and 6 present legal regulations in Poland and other countries concerning permissible concentration of heavy metals entering the environment together with sewage sludge during its agricultural application.

Table 5

Permissible heavy metals content [mg · kg$^{-1}$ d.m.] in sewage sludge for agriculture disposal

| Metals | Poland | UE directive | Denmark | Austria | Germany | Switzer-land | Sweden | USA |
|---|---|---|---|---|---|---|---|---|
| Pb | 750 | 750 | 120 | 500 | 900 | 500 | 100 | 840 |
| Cd | 20 | 20 | 0.8 | 10 | 10 | 5 | 2 | 85 |
| Cr | 500 | 1000 | 100 | 500 | 900 | 500 | 100 | 3000 |
| Cu | 1000 | 1000 | 1000 | 500 | 800 | 600 | 600 | 4300 |
| Ni | 300 | 300 | 30 | 100 | 200 | 80 | 50 | 420 |
| Hg | 16 | 16 | 0.8 | 10 | 8 | 5 | 25 | 57 |
| Zn | 2500 | 2500 | 4000 | 2000 | 2500 | 2000 | 800 | 7500 |

Source: own data processing [7–9].

**Working document on sludge. 3rd Draft. Brussels, 27 April 2000** – a document presenting significant tightening of quality expectations and limitations in agricultural application of sludge. Detailed plans were to decrease total amount of potentially dangerous substances and elements [10]. Council Directive 86/278/EEC of June 1986 on protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture, was replaced with Council Directive 91/962/EWG and Council Regulation 807/2003/WE (Dz. Urz. WE L 181 from 04.07.1986, page 6, with later changes). At present, European Commission is elaborating on "Working document sludge and biowaste" 21st September 2010, Brussels, in which same changes in Sewage sludge Directive are suggested.

Table 6 presents permissible heavy metal content in sludge applied in agriculture.

Table 6

EU heavy metals limits in sewage sludge applied in agriculture

| Metal | Heavy metals limits [$mg \cdot kg^{-1}$ d.m.] | | |
|---|---|---|---|
| | 1986/278/EWG | Working dokument on sludge. 3rd Draft | Working document sludge and biowaste 21 September 2010, Brussels |
| Cd | 20–40 | 10 | 10 |
| Cr | — | 1000 | 1000 |
| Cu | 1000–1750 | 1000 | 1000 |
| Hg | 16–25 | 10 | 10 |
| Ni | 300–400 | 300 | 300 |
| Pb | 750–1200 | 750 | 500 |
| Zn | 2500–4000 | 2500 | 2500 |

Source: [10–12].

Analysis of Tables 5 and 6 points out a few important changes in regulations concerning sludge agricultural application in Poland and other countries of EU. Update of Sewage Sludge Regulation in 2010 has changed mostly attitude to the issue of heavy metals doses entering the environment. Maximal content of heavy metals in sewage sludge and admissible levels of sludge doses allowed within year per hectare were defined.

Suggested changes relate to:

– Requirements for pollutants prevention,

– Sewage sludge treatment and application as well as conditions of its agricultural application.

Changes regarding sewage sludge utilization on soil are as follow:

– Implementation of stricter solution for heavy metals,

– Standards determination for organic compounds and pathogenic organisms,

– Higher requirements concerning sewage sludge application, sampling and monitoring.

Planned changes in Sewage Sludge Directive do not consider:

– Implementation of strict standards for all substances and prohibition of sludge application on some crops, for the reason of too high costs,

– Prohibition of sludge application on soil.

Changes in Sewage Sludge Directive drive to reduction of dangerous organic substances and heavy metals in sewage discharged to sewerage, which enter sewage sludge. Regulation changes in the range of sewage sludge concern implementation of sewage sludge producer responsibility, certification and requirement applied information for produces and sludge receiver.

## Conclusions

Analysis of presented results unequivocally shows low content of heavy metals entering the environment as a result of agricultural application of sludge from dairy sewage treatment. Discovered amounts of heavy metals in the sludge are very often several or a few hundred times lower than permissible ones according to various documents and legal acts in Poland and worldwide. At the same time examinations have shown very low content of heavy metals in structural materials applied during the process of windrow composting or with the use of California earthworm (*Eisenia fetida*) [13, 14]. Adding structural materials has positively influenced these characteristics of treated sludge which are significant in environmental application. The process of composting and vermicomposting has changed sludge structure and additionally enriched it with fertilizer values (N, P, K content) and soil formation values (eg humification and mineralization process).

These are technologies which enrich sewage sludge by causing quick fading of putrid smell, formation of permanent crumb texture which enhances plant growth or increase in level of nutrients on utilized base [15–17].

Examined sludge from sewage treatment plant in Wysokie Mazowieckie fulfills basic criteria and can be used untreated for agricultural and non-agricultural purposes and for soil remediation.

Until now, Polish research on agricultural application of sewage sludge has not attempted to test properly prepared dairy sludge. However, managing sludge from dairy sewage treatment plants in large production facilities is highly problematic. Different character and physical and chemical composition of dairy sludge in comparison with typical municipal sludge (among others much higher nitrogen and phosphorus content in sludge in relation to carbon compounds, and presence of large amounts of coagulated protein) are the reason why technological methods and parameters of sludge treatment developed for municipal sewage sludge, eg into compost, do not bring the desired effects [18, 19]. At the same time, according to Waste Management Act sludge from dairy sewage treatment plants is considered as municipal sewage sludge [20].

Both Polish and international examinations of production of compost, vermicompost, remediation measures, and natural fertilizers mostly concern municipal sludge of different chemistry and character than dairy sludge.

1. Suggested legislative changes in Poland and EU concerning environmental sewage sludge application prefer application of sewage sludge with low heavy metals content and high nutrient values, as well as sanitary safe. Research has demonstrated that sludge from dairy sewage treatment plants meet these requirements.

2. Sludge from dairy sewage treatment plants, which constitutes 40 % of sewage sludge generated in Podlasie province, can be significant source of nutrients in soil.

3. Composting process of the sludge from dairy sewage treatment plants increases its value for agricultural application.

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## METALE CIĘŻKIE I SKŁADNIKI POKARMOWE WPROWADZANE DO ŚRODOWISKA PRZYRODNICZEGO Z OSADAMI ŚCIEKOWYMI


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**Abstrakt:** W artykule przedstawiono wyniki badań osadów z oczyszczalni ścieków mleczarskich w północno-wschodniej Polsce oraz wyniki badań kompostów uzyskanych w instalacjach badawczych stosujących niskonakładowe metody przetwarzania osadów ściekowych mleczarskich oraz dodawanych do tych osadów materiałów strukturotwórczych. Dokonano analizy substancji chemicznych i oceny ich ilości z wprowadzanymi do środowiska osadami mleczarskimi. Odniesiono się do ilości dopuszczalnych metali ciężkich zawartych w osadach w prawodawstwie polskim i w innych krajach. Stwierdzono bardzo małą szkodliwość osadów mleczarskich. Wykazano, że proces kompostowania i wermikompostowania zmienił strukturę osadu, a także wzbogacił dodatkowo o wartości nawozowe (zawartość N, P, K) i glebotwórcze.

**Słowa kluczowe:** wermikultura, osady ściekowe, niskonakładowe metody, kompost, wermikompost, przemysł mleczarski