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THE USE OF PLANTS FOR PURIFICATION OF WASTEWATER FROM PHARMACEUTICAL FACTORIES

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Abstract. Based on literary analysis, the effectiveness of a range of plants (aquatic: *Lemna aoukikusa*, *Lemna minor*, *Spirodela polyrhiza*, *Lemna aequinoctialis*; vetiver grass *Chrysopogon zizanioides*) for the purification of wastewater from antibiotics has been investigated. It has been found that the removal efficiency for various types of antibiotics and their concentrations reaches 70 percent or more. This suggests the potential application of these aquatic plants for phytoremediation of wastewater containing antibiotic contaminants.

Keywords: phytoremediation, wastewater, pharmaceutical companies, pharmaceuticals, antibiotics, plants.

1. Introduction

The purification of wastewater generated by pharmaceutical factories is considered a significant challenge, as they constitute a substantial source of diverse pollutants. Wastewater from pharmaceutical enterprises may contain various substances depending on the processes occurring within these facilities. The main pollutants can include:

1. Pharmaceutical compounds: various chemical compounds used in the production of medicines may find their way into wastewater through manufacturing and equipment cleaning processes.
2. Solvents and reagents: pharmaceutical production often involves the use of different solvents and chemical reagents that can enter wastewater.
3. Equipment and process-related contaminants: lubricants, oils, equipment waste, and other substances may inadvertently enter wastewater during technological processes.
4. Organic contaminants: substances of organic origin, such as biological residues, bacteria, and other microorganisms, may also be present in wastewater.

In particular, antibiotics play a particularly important role as they contribute to the development of bacterial resistance to existing drugs, posing a threat to the effective control of infectious diseases. Moreover, not all remnants of antibiotics are metabolized in the human body after use, and a significant portion of them ends up in wastewater. These biologically active compounds are difficult to remove using traditional purification methods employed by municipal treatment facilities.

Information exists about the presence of tetracycline antibiotics in treated wastewater at a concentration of 652.6 ng/dm^3 and sulfonamide antibiotics at 261.1 ng/dm^3 (Jendrzejewska, Karwowska, 2018).

Research on the impact of antibiotics from different classes (doxycycline, gentamicin, penicillin, nitrofurantoin, and rifampicin) within the concentration range of 100–300 μg/dm^3 indicates a significant proliferation of antibiotic-resistant microorganisms in the activated sludge. The highest number of bacteria

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became resistant to the following antibiotics: nitrofurantoin and penicillin (Gao et al., 2012).

Therefore, treatment facilities are considered to contribute to the evolution and spread of antibiotic-resistant strains.

Hence, the development, improvement, and implementation of effective methods and technologies for purifying wastewater from pharmaceutical enterprises are pressing issues. The main objective of this study is to explore the literature concerning the efficacy of using aquatic plants for the removal of pharmaceutical residues from wastewater produced by pharmaceutical companies.

2. Theoretical part

The purification of wastewater from antibiotic contamination using plants falls under the ecological biotechnology known as phytosanitation or phytoremediation. This method utilizes plants to remove or reduce pollutants, such as antibiotics, from aquatic environments. The fundamental concept is that plants can absorb pollutants through their diverse root systems.

The process of phytoremediation for antibiotic-contaminated wastewater may involve the following stages (Singh, Pant, 2023; Ansari et al., 2020):

1. Phytoextraction: Plants are cultivated in contaminated water bodies or containers, where they absorb antibiotics through their roots. Once the plants have absorbed antibiotics, they are harvested and disposed of.
2. Phytostimulation: Plants help activate microorganisms in the soil, which, in turn, break down antibiotics into less hazardous compounds.
3. Phytodegradation: Plants can break down or degrade antibiotics in water or soil through chemical or biological processes.

The process of phytoremediation is complex and depends on various factors, such as the type of antibiotics, their concentrations, characteristics of the aquatic environments, and the species of plants used. Some plants known to be used in antibiotic phytoremediation (Ansari et al., 2020; Chugh et al., 2020) include:

1. Lettuce (*Lactuca sativa*) – This plant is known for its ability to phytoextract, accumulating antibiotics in its tissues through its roots. These plants can then be harvested and disposed of.
2. Algae (*Algae*) – Certain types of algae can facilitate antibiotic removal from water through phytodegradation.
3. Rapeseed (*Brassica napus*) – This plant can be used for antibiotic phytoextraction, particularly in cases where the root system has a large surface area for absorption.
4. Reed (*Phragmites*) – It is known for its ability to collect and retain antibiotics in its roots and stems.
5. Water Lily (*Nymphaea spp.*) – This plant can aid in water purification from antibiotics through phytostimulation and phytodegradation.

It's important to note that the effectiveness of phytoremediation may depend on a range of factors, including climatic conditions, pollution levels, soil type, and many others. Research in this field is ongoing, and new approaches and plant species for antibiotic-contaminated wastewater phytoremediation continue to be explored.

Purification of wastewater from antibiotics using algae is a phytosanitation method that harnesses the natural potential of algae to remove antibiotics and other pollutants from aquatic environments. Algae are excellent candidates for this process due to their large surface area for absorption and their ability to biologically degrade various compounds, including antibiotics.

The main mechanisms through which algae can purify wastewater from antibiotics include (Dhir, 2013; Mccutcheon, Schnoor, 2004):

1. Phytoextraction: Algae can actively accumulate antibiotics within their cells or on their body surface through phytoextraction. When grown in contaminated water bodies, algae absorb antibiotics from the surrounding environment.
2. Biological degradation: Certain types of algae can biologically degrade antibiotics by producing enzymes or other biochemical processes. This mechanism breaks down antibiotics into less toxic or harmful compounds.
3. Adsorption: Algae can also physically adsorb antibiotics onto their surface, removing them from the water. This can occur through adsorption on cell walls or surface projections of the algae.
4. Root system uptake: In some cases, algae with root systems can absorb antibiotics through their roots, which can then be removed along with the plants.

For effective phytoremediation using algae, it is important to select appropriate algae species that possess the necessary characteristics for treating specific types of antibiotics and pollutants. Environmental parameters such as pH, temperature, antibiotic concentration, and others should also be considered to provide optimal conditions for algae growth and ensure the successful progression of the phytoremediation process.

Wastewater purification from antibiotics using algae is considered an ecologically sound method, known as phytoremediation. This approach harnesses the natural capacity of algae to remove antibiotics and other pollutants from aquatic environments. Algae, due to their expansive surface area and ability for biological degradation, are prime candidates for this process.

One such candidate is the duckweed, which is often considered for phytoremediation of antibiotic-contaminated wastewater. Duckweeds are small floating higher aquatic plants with a green hue, belonging to the *Lemnaceae* family. This family comprises five different genera: *Landoltia*, *Lemna*, *Spirodella*, *Wolffia*, and *Wolffiella* (Ali et al., 2016). Duckweed demonstrates significant potential for absorbing pollutants, making it an effective, environmentally friendly, and cost-efficient means of wastewater treatment.

Scientific studies (Habaki et al., 2023) confirm the high efficiency of using *Lemna aoukikusa*, a common species of duckweed depicted in Fig. 1, for treating antibiotic-containing wastewater. Experiments demonstrated that duckweed can thrive in wastewater containing antibiotics such as ciprofloxacin and sulfamethoxazole, even at high concentrations, while simultaneously absorbing these antibiotics from the aqueous solution. This applies even to cases of heavy wastewater contamination, with initial antibiotic concentrations reaching $5.0 \cdot 10^{-2}$ mol·m^{-3}.

In a separate investigation (Balarak, 2017), the removal of ciprofloxacin from water solutions through adsorption using activated charcoal produced from the floating water fern *Azolla filiculoides* was studied. This water fern, shown in Fig. 2, exhibits rapid growth and is the largest species within the *Azolla* genus. Research results indicated that at the antibiotic's concentration of 10 mg/dm^3 in water, using a dosage of 2.5 g/dm^3 of activated charcoal and a contact time of 75 minutes, the removal of the antibiotic was 99.1 %. Thus, it is evident that activated charcoal produced from the floating water fern *Azolla filiculoides* can be successfully utilized for effective removal of antibiotics from wastewater.

Other studies (Gomes et al., 2020; Malovanyy et al., 2021; Malovanyy et al., 2018; Soloviy et al., 2019) on water purification from antibiotics indicate that the aquatic macrophyte *Lemna minor* can accumulate significant concentrations of certain antibiotics (such as amoxicillin, enrofloxacin, and oxytetracycline) from the aquatic environment, even when they are present simultaneously at concentrations of amoxicillin 2 μg/dm^3, enrofloxacin 2 μg/dm^3, and oxytetracycline 1 μg/dm^3. This highlights the potential use of such aquatic plants for phytoremediation of natural water bodies that may be contaminated with these antibiotics. This particular plant is depicted in Fig. 3.

Fig. 1. Photo of the duckweed species *Lemna aoukikusa* (Habaki et al., 2023)

Fig. 2. Photo of the aquatic fern *Azolla filiculoides* (Balarak, 2017)

In the course of the investigation (Singh et al., 2018), an assessment of the potential toxicity of amoxicillin to the *Spirodela polyrhiza* duckweed, the largest species within the genus *Spirodela*, was conducted (Fig. 4). This perennial plant floats on the water's surface and possesses root bundles. Over a period of 7 days, *Spirodela polyrhiza* was exposed to amoxicillin at low (0.0001–0.01 mg/dm^3) and high (0.1–1 mg/dm^3) concentrations. It was observed that the antibiotic had a toxic impact on *Spirodela polyrhiza*, even at low concentrations. Nevertheless, despite this toxicity, the duckweed contributed to antibiotic degradation in the water throughout the entire phytoremediation process.

Fig. 3. Photo of the aquatic macrophyte *Lemna minor* (Gomes et al., 2020)

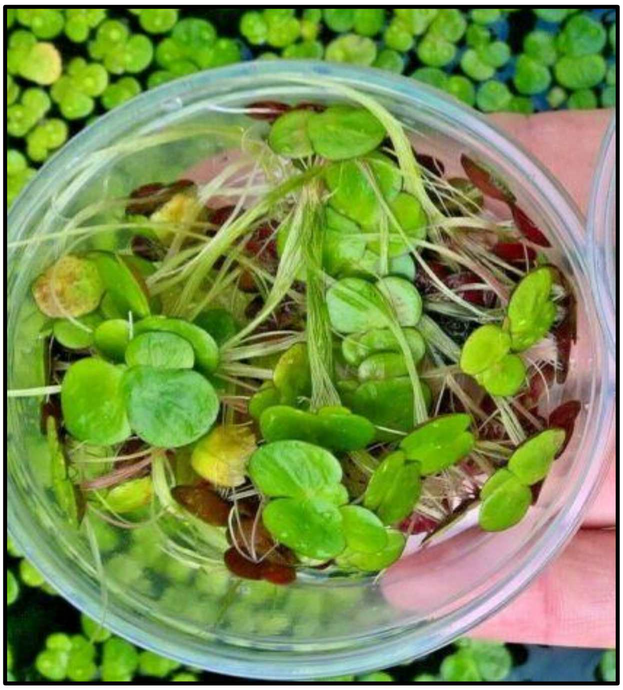

Fig. 4. Photo of the duckweed *Spirodela polyrhiza* (Singh et al., 2018)

The study (Huang et al., 2022) allowed for the investigation of streptomycin's impact on physiological changes and the absorption capacity of *Lemna aequinoctialis*, a species of duckweed that has an annual growth cycle (Fig. 5). *Lemna aequinoctialis* was exposed to varying concentrations of streptomycin ranging from 0.1 to 10 mmol/dm^3 for different durations: 0, 5, 10, 15, and 20 days. The results indicate that high concentrations of streptomycin exceeding 1 mmol/dm^3 led to a reduction in duckweed biomass by 21.5–41.5 %. A significant decrease in streptomycin content – 72–82 % over the course of 20 days compared to the reference group without duckweed (40–55 %) – was observed. This suggests the high capability of *Lemna aequinoctialis* to remove streptomycin. Therefore, the findings confirm that this duckweed species can contribute to the degradation of streptomycin and can be utilized for wastewater purification purposes.

Fig. 5. Photo of the duckweed *Lemna aequinoctialis* (Huang et al., 2022)

A study (Panja et al., 2020) was conducted to explore the potential of using the perennial grass *Chrysopogon zizanioides*, commonly known as vetiver, for the removal of antibiotics – ciprofloxacin and tetracycline – from wastewater. Vetiver is a fast-growing plant with long vertical roots that can reach up to 4 meters in length and is capable of growing hydroponically (Fig. 6). Over a 30-day period, significant removal of antibiotics and biogenic compounds (N and P compounds) from wastewater was observed using vetiver. The plant removed over 90 % of antibiotics and effectively removed nitrates (>40 %), phosphates (>60 %), total organic carbon (>50 %), and also reduced chemical oxygen demand (COD) by more than 40 %.

For antibiotic removal in the study (Maldonado et al., 2022), various methods were employed, including phytoremediation using floating aquatic plants such as duckweed and water fern, with successful outcomes. This study examines the efficiency of removing pharmaceuticals, particularly antibiotics, through the use of *Lemna* and *Azolla*, aiming for a better understanding of the physiological aspects of phytoremediation. Certain aspects were analyzed, including plant physiological processes in the presence of these pollutants and their phytotoxic consequences at high concentrations. The initiation of the metabolism of toxic compounds involves the absorption of antibiotics, culminating in the sequestration of assimilated compounds in vacuoles, apoplast, and cell walls. Hence, plants contribute to the removal of toxic compounds from water.

Fig. 6. Photo of the perennial grass vetiver *Chrysopogon zizanioides* (Panja et al., 2020)

3. Conclusions

Analysis of methods and technologies for the purification of wastewater from antibiotics using a range of plants: aquatic plants such as duckweeds *Lemna aoukikusa*, *Lemna minor*, *Spirodela polyrhiza*, *Lemna aequinoctialis*; and vetiver grass *Chrysopogon zizanioides*, demonstrated their high effectiveness against various antibiotics. It was found that these plants exhibit significant efficacy across different types of antibiotics. However, a crucial factor influencing removal efficiency is the type of antibiotic and its initial concentration in wastewater.

The research was conducted for specific types of antibiotics and within certain concentration ranges, thus the obtained data can only be applied under analogous specific conditions. Therefore, it is necessary to individually investigate the technological regime of wastewater treatment for each pharmaceutical enterprise targeting a specific type or group of antibiotics using plants, taking into account the specific composition of the wastewater.

Hence, establishing correlations between the parameters of the technological purification process using aquatic plants and the characteristics of wastewater from a particular pharmaceutical company is the primary goal of further research in this field.

References

Ali, Z., Waheed, H., G. Kazi, A., Hayat, A., & Ahmad, M. (2016). Chapter 16 – Duckweed: An Efficient Hyperaccumulator of Heavy Metals in Water Bodies. *Plant Metal Interaction*, *2016*, 411–429. doi: https://doi.org/10.1016/B978-0-12-803158-2.00016-3

Ansari, A. A., Naeem M., Gill, S. S., & AlZuaibr, F. M. (2020). Phytoremediation of contaminated waters: an eco-friendly technology based on aquatic macrophytes application. *The Egyptian Journal of Aquatic Research*, *46*(4), 371–376. doi: https://doi.org/10.1016/j.ejar.2020.03.002

Balarak, D., Mostafapour, F. K., Akbari, H., & Joghtaei, A. (2017). Adsorption of amoxicillin antibiotic from pharmaceutical wastewater by activated carbon prepared from Azolla filiculoides. *Journal of Pharmaceutical Research International*, *18*(3), 1–13. doi: http://dx.doi.org/10.9734/JPRI/2017/35607

Dhir, B. (2013). *Phytoremediation: Role of Aquatic Plants in Environmental Clean-Up*. Springer New Delhi. doi: https://doi.org/10.1007/978-81-322-1307-9

Chugh, M., Kumar, L., P Shah, Maulin, & Bharadvaja, N. (2022). Algal Bioremediation of heavy metals: An insight into removal mechanisms, recovery of by-products, challenges, and future opportunities. *Energy Nexus*, 7, 10129. doi: https://doi.org/10.1016/j.nexus.2022.100129

Gomes, M. P., Moreira Brito J. C., Rocha D., C., Navarro-Silva, M. A., & Juneau, P. (2020). Individual and combined effects of amoxicillin, enrofloxacin, and oxytetracycline on *Lemna minor* physiology. *Ecotoxicology and Environmental Safety, Elsevier*, *203*, 11025. doi: https://doi.org/10.1016/j.ecoenv.2020.111025

Gao, P., Munir, M., & Xagoraraki, I. (2012). Correlation of tetracycline and sulfonamide antibiotics with corresponding resistance genes and resistant bacteria in a conventional municipal wastewater treatment plant. *Science of The Total Environment*, *421–422*, 173–183. doi: https://doi.org/10.1016/j.scitotenv.2012.01.061

Jendrzejewska, N., & Karwowska, E. (2018). The influence of antibiotics on wastewater treatment processes and the development of antibiotic-resistant bacteria. *Water Science and Technology*, *77*(9), 2320–2326. doi: 1https://doi.org/10.2166/wst.2018.153

Habaki, H., Thyagarajan, N., Li, Z., Wang, S., Zhang, J., & Egashira, R. (2023). Removal of antibiotics from pharmaceutical wastewater using *Lemna Aoukikusa* (duckweed). *Separation Science and Technology*, *58*, 1491–1501. doi: https://doi: 10.1080/01496395.2023.2195544

Huang, W., & Kong, R., & Chen, L., An, Y. (2022). Physiological responses and antibiotic-degradation capacity of duckweed (*Lemna 1aequinoctialis*) exposed to streptomycin. *Frontiers in Plant Science*, *13*. doi: https://doi.org/10.3389/fpls.2022.1065199

Maldonado, I., G. Moreno Terrazas, E., & Zirena Vilca, F. (2022). Application of duckweed (*Lemna* sp.) and water fern (*Azolla* sp.) in the removal of pharmaceutical residues in water: State of art focus on antibiotics. *Science of The Total Environment*, *838,* 156565. doi: https://doi.org/10.1016/j.scitotenv.2022.156565

Malovanyy, M. S., Soloviy, Kh. M., & Nykyforov, V. V. (2018). Conditions for development and cultivation of cyanobacteria for multi-target application (literature review). *Environmental Problems*, *3*(1), 1–11.

Malovanyy, M., Tymchuk, I., Balandiukh, Iu., Soloviy, Kh., Zhuk, V., Kopiy, M., Stokalyuk, O., & Petrushka, K. (2021). Optimum collection and concentration strategies of hydrobionts excess biomass in biological surface water purifying technologies. *Environmental Problems*, *6*(1), 40–47. doi: https://doi.org/10.23939/ep2021.01.040

Mccutcheon, S., & Schnoor, J. (2004). Phytoremediation Transformation and Control of Contaminants. *Environmental Science and Pollution Research*, *11*, 40. doi: https://doi.org/10.1007/BF02980279

Panja, S., Sarkar, D., & Datta, R. (2020). Removal of antibiotics and nutrients by Vetiver grass (*Chrysopogon zizanioides*) from secondary wastewater effluent. *International Journal of Phytoremediation*, *22*, 764–773. doi: https://doi.org/10.1080/15226514.2019.1710813

Singh, V., Pandey, B., & Suthar, S. (2018). Phytotoxicity of amoxicillin to the duckweed *Spirodela polyrhiza*: Growth, oxidative stress, biochemical traits and antibiotic degradation. *Chemosphere*, *201*, 492–502. doi: https://doi.org/10.1016/j.chemosphere.2018.03.010

Singh, H., & Pant, G. (2023). Phytoremediation: Low input-based ecological approach for sustainable environment. *Applied Water Science*, *13*. doi: http://dx.doi.org/10.1007/s13201-023-01898-2

Soloviy, Kh., Malovanyy, M. (2019). Freshwater Ecosystem Macrophytes and Microphytes: Development, Environmental Problems, Usage as Raw Material. Review. *Environmental Problems*, *4*(3), 115–124. doi: https://doi.org/10.23939/ep2019.03.115