Safe Reuse of Landfill Leachates for Irrigation Purposes

ISARI A. E.[1], GRILLA E.[1], KOKKINOS P.[1,*], PARTHENIDIS P.[2,3], EVGENIDOU E.[2,3], LAMBROPOULOU D.[2,3], KALAVROUZIOTIS I. K.[1]

[1]Laboratory of Sustainable Waste Management Technologies,
School of Science and Technology,
Hellenic Open University,
Parodos Aristotelous 18, 26335, Patras,
GREECE

[2]Laboratory of Environmental Pollution Control,
School of Chemistry, Faculty of Sciences,
Aristotle University of Thessaloniki,
GREECE

[3]Centre for Interdisciplinary Research and Innovation (CIRI-AUTH),
Balkan Center, Thessaloniki,
10th km Thessaloniki-Thermi Rd, GR 57001,
GREECE

Corresponding Author

Abstract: - The purpose of the present study was the assessment of the potential reusability of landfill leachates for agricultural irrigation, after advanced oxidation treatments. The UV/Fenton and Fenton processes were applied to the treatment of two different landfill leachate samples, L1 and L2, classified as intermediates, collected from two different landfills located in Macedonia, Greece. Samples were characterized by high COD and TOC values, ranging from 5500-6100 mg/l and 1700-1780 mg/l, respectively. The treatment efficacy and toxicity characteristics were evaluated by conducting phytotoxicity tests. *Sorghum saccharatum* seeds were used and the germination index (GI) was estimated. The results showed that the UV/Fenton process achieved better organic matter removal rates for both samples. The untreated undiluted leachates L1 and L2 were highly toxic, resulting in GI values of 0. The results of the study showed that *Sorghum saccharatum* seed germination depends on the nature of the irrigation media and that optimum germination rates were achieved at dilutions of treated leachates with ratios higher than 1:4.

Key-Words: - landfill leachate, phytotoxicity, advanced oxidation process, UV/Fenton

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1 Introduction

Landfill leachate is a dark-coloured complex and highly variable wastewater that is produced when rainwater percolates through waste materials disposed of in a landfill, [1]. Due to its composition, it is one of the major sources of pollution if discharged into an environmental system, [2]. It typically contains a variety of micropollutants, including phenols, plasticizers, pharmaceuticals, pesticides, personal care products (PCPs), endocrine disruptors, PCBs, PAHs and organo-chlorinated compounds as recalcitrant substances, refractory compounds, heavy metals, [3], [4], [5], inorganic compounds and pathogens, [3]. Therefore, it is characterized by high levels of organic load expressed as TOC, COD, BOD_5, and inorganic load as salinity (Na^+, K^+, Ca^{2+} or Cl^- ions), ammonia, [2], [5] and Cr, Cd, Pb, Hg, Ni, Cu, Zn, Fe, and Se, [6].

A common pollutant indicator in landfill leachate is the colour, which mainly comes from the decomposition of organic compounds, such as humic acid, which may turn the water's colour yellow or dark brown. Total dissolved solids (TDS) play a key role, as they display the influence of certain cations and anions, on wastewater. However, landfill leachate's composition and quantity differ depending on the age, ambient air temperature, permeability, degradation procedure, climate, and type of waste. It is known that as landfill leachate ages, the pH increases, [7], the organic load decreases and stabilizes, [5], and the metal concentration becomes lower, but it still remains hazardous waste that requires proper treatment to minimize the risks to the ground, surface water, soil, animal and human health, [7].

Existing landfill leachate treatment technologies can be classified into two main categories: conventional and advanced. Conventional methods considered as the combined treatment of domestic sewage with landfill leachate, recycling, biodegradation processes (aerobic and anaerobic treatment), and physicochemical methods, such as air stripping, flotation, coagulation-flocculation, settling, chemical precipitation, adsorption, and their combination, [8], [9], [10], [11]. Advanced methods are a broad category of processes that includes various filtration methods such as ultrafiltration, microfiltration, nanofiltration, and reverse osmosis, as well as advanced oxidation processes (AOPs). Each treatment has its pros and cons. For example, biological processes can be very effective in removing organic matter and total N from "young" leachates when their BOD/COD ratio is above 0.5, but over time the presence of humic and fulvic acids as indicators of leachates' maturity, tends to limit the effectiveness of the method, [9]. Also, the typical coagulation-flocculation process can be used as the pre-treatment or final step for the removal of non-biodegradable organic matter. However, the use of chemical reagents, the production of a constant volume of sludge, and the noticeable increase in the concentration of aluminum or iron in the liquid phase do not make the application of the method ideal, [9]. Thus, based on environmental and economic criteria, it is essential to choose green technologies, which aim to minimize chemical and energy consumption, produce less sludge and by-products, and can degrade a wide range of micropollutants, [12].

AOPs can be considered "cleaner" technologies with great abatement effectiveness on toxic substances. AOPs are based on the production of highly reactive non-selective oxygen species (ROS), which cause redox reactions, [12]. Electrochemical oxidation, electrocoagulation, photocatalysis activated by semiconductors, sonolysis, ozonation, Fenton, photo-Fenton and UV/Fenton are some of the AOPs, [12], [13]. Among them, the Fenton process, a reaction between dissolved iron ions (Fe^{2+}) and hydrogen peroxide (H_2O_2), resulting in the production of hydroxyl radicals, leads to the formation of oxidative radicals, which react with the organic pollutants in the sample, causing their degradation or transformation. The Fenton process can be combined with other AOPs, like UV-light, where the synergistic effect of different processes can significantly improve the oxidation efficiency of contaminants compared with the individual treatment technology, [14]. At the UV/Fenton process, H_2O_2 is catalysed by Fe^{2+} producing more OH radicals. Simultaneously, Fe^{3+} can be transformed to Fe^{2+} by UV, to catalyse H_2O_2, [15]. The main advantages of Fenton and UV/Fenton processes are the high reaction rate, the high efficiency in degrading pollutants, and mineralizing landfill leachates, [1]. Thus, treated landfill leachates present an opportunity not only for their safe disposal in water receivers but also for their use as a potential source of water for agricultural purposes, such as irrigation, soil amendment and fertigation.

Reusing leachate in agriculture can be a challenging task as it requires careful treatment and management to ensure that it is not toxic to the environment. Therefore, ecotoxicological tests should be performed to indicate the impact on living organisms. Ecotoxicity tests show a wide variety. Some of these are based on the use of plants, and called phytotoxicity tests. These tests are mainly based on the assessment of germination rate, root growth and/or plant growth rate, [16].

The aim of the present research, which was carried out as part of the UV-LEACH project, was to evaluate the potential of reusing leachates for agricultural irrigation purposes, after their treatment with AOPs (Fenton and UV/Fenton process), and the evaluation of their phytotoxicity, using *Sorghum saccharatum* seeds.

2 Problem Formulation

The agricultural sector is one of the greatest users of freshwater, accounting for approximately 50% of total annual water consumption, [17]. In May 2018, the European Commission proposed a regulation, which sets new standards that reclaimed water must meet to be used for agricultural irrigation, alleviating water stress, [18], followed by the Water Reuse Regulation ((EU)2020/741). In 2050, the world population is estimated to reach around 9 billion. Consequently, water scarcity is an actual threat to food security because of the limited water resources.

The necessity of safe water reuse drew the attention of the research community to the proper treatment of landfill leachates. For this reason, many studies focused on new advanced oxidation processes for the elimination of emerging contaminants.

In the present study, two leachate samples, L1 and L2, were collected from two different landfills located in Macedonia (Greece). The samples can be characterized as intermediates, based on the typical classification of leachates, [19]. Their COD and TOC values were determined and found to be particularly high, ranging from 5500-6100 mg/l and 1700-1780 mg/l, respectively. Therefore, their organic load makes them unsuitable for reuse, without proper treatment.

3 Problem Solution

Samples L1 and L2 were treated by Fenton and UV/Fenton processes. Specifically, the experimental procedure started with the preparation of each sample with pH adjustment at 3 using sulfuric acid (H_2SO_4). A sample volume of 300 ml was introduced into the reactor. H_2O_2 and Fe(II) were added in the ratio of COD: $[H_2O_2]$ = 1:1 and [Fe(II)]: $[H_2O_2]$ = 1:10, respectively. The UV lamp was placed in the reactor at t = 0 min. After 60 minutes of treatment, the samples were centrifuged and filtered (0.45 μm filter). The treated samples were subjected to analysis to evaluate the homogeneous oxidative processes for their efficiency. Under the applied experimental conditions, the UV/Fenton process achieved higher COD and TOC removal for landfill leachates L1 and L2 (Table 1), highlighting it as the optimal organic load removal technique. The differences noted in the removal rates are probably due to the different characteristics of the leachates such as their age and the type of pollutants contained, [20]. It should be mentioned that a part of the organic contaminants is not degraded, and probably is transferred to another phase through sludge formation. The Fenton process can remove an amount of organic matter via coagulation that occurs due to the iron salt addition in combination with the acidic pH of the solution, [21].

Despite the improvement of leachate characteristics, their reuse for irrigation purposes should be considered under preliminary phytotoxicity experiments at the lab scale. The evaluation of toxicity of untreated and treated samples was conducted by phytotoxicity determination using higher plant species, such as *Sorghum saccharatum*, (Figure 2). All tests were carried out with Plant Growth Incubator (MRC, PGI-550RH). Distilled water was used for positive control. Treated leachate samples were diluted with distilled water at dilution ratios of 1:2, 1:4, 1:6, 1:10, 1:25, 1:50, 1:75 and 1:100. Four (4) ml of each concentrated and diluted leachate sample was added to a Petri dish containing four (4) pieces of blotting paper onto which 20 seeds were evenly placed. Petri dishes were incubated for 72 h at 25 ºC. After the incubation period, sprouted seeds were counted and the germination index (GI) was calculated.

As can be seen in Figure 1, there is a notable difference in seed germination between the concentrated untreated and treated leachates. The absence of seed germination, corresponding to zero values of GI for concentrated L1 and L2 samples, reveals the high level of their toxicity and underlines the need for their effective treatment, [22]. Notably, even a dilution ratio of 1:2 of the untreated samples cannot result in any seed germination. However, a dilution ratio of 1:4 may result in seed germination since GI increases to 55-60 %. A percentage that successively increases as the dilution factor increases (GI ranges from 70-90%). For leachate sample L2, the oxidation processes achieved similar levels of GI, i.e. 65 % for the UV/Fenton process, and 75 % for the Fenton process, showing that none of the applied treatments may be considered optimal. On the contrary, a treated L1 sample may cause seed germination only after dilution. GI values ranged between 80-95% for both samples after dilution. Treatment of landfill leachate L1 with both processes resulted in lower GI values compared to L2.

Table 1. Percent removal of leachate samples after Fenton and UV/Fenton treatment.

Physicochemical Characteristics	L1		L2	
	Fenton	UV/Fenton	Fenton	UV/Fenton
COD removal (%)	32.17	52.5	45.67	54.62
TOC removal (%)	57	67.47	69.49	74.2

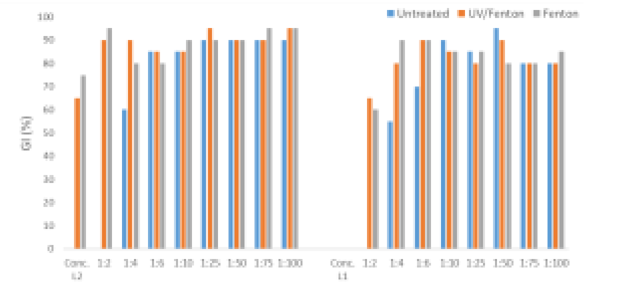

Fig. 1: Germination Index (GI) of untreated, Fenton and UV/Fenton treated non-diluted and diluted leachate samples L1 and L2

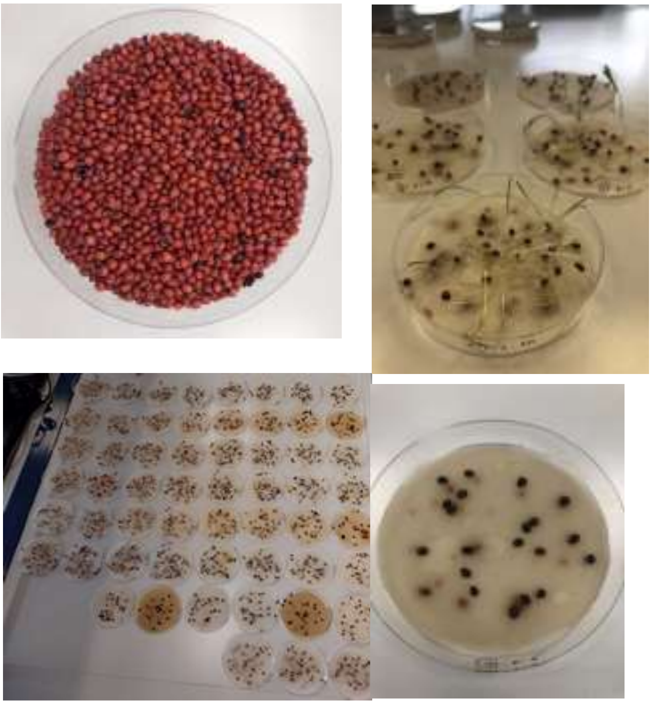

Fig. 2: Seeds of *Sorghum saccharatum* (upper left), germinated seeds (upper right), experimental layout for GI evaluation using seeds of *Sorghum saccharatum* and different dilutions of leachate samples, including positive and negative controls (lower left), and a Petri dish with no seeds germinated (lower right).

In a more general context, the phytotoxicity results indicate the need to dilute the samples to a ratio greater than 1:4, after they have first been treated so that the germination of the seeds of the selected plant species is possible. This result is consistent with studies reporting that dilution decreases the organic, and inorganic load and electric conductivity of the leachates resulting in not only high rates of germination but also high productivity of selective plant species, [23], [24]. Regarding the effect of advanced oxidation processes on the germination index, it is evident that the differences are negligible, with higher percentages presented by sample L2, as it also had the highest removal of organic load during its treatment.

4 Conclusion

The experimental results of this preliminary study revealed that the Fenton process enhanced by UV radiation can be a promising treatment for leachates. In addition, the results of the phytotoxicity tests are an important indication that the treated leachates with appropriate dilution can be used for agricultural irrigation, being at the same time a way out of water scarcity and a potential source of nutrients for soil fertilization.

Further work is needed to optimize landfill leachate treatment processes to minimize potential risks from their use as a water source. In this context, phytotoxicity tests should be carried out with different plant seeds, which vary according to their susceptibility to toxicity. A future study should also be to irrigate plants under greenhouse conditions to assess the effect of treated leachate irrigation on plant growth, soil quality and potential groundwater contamination.

Finally, the present study is the starting point for the treatment of very difficult liquid waste and its reuse according to the terms of sustainability and the circular economy. Its results at the laboratory scale can be applied in practice with a view to the future solution to the problem of increasing pollution and water scarcity.

References:

[1] Parthenidis, P., Evgenidou, E., & Lambropoulou, D. (2022). Wet and supercritical oxidation for landfill leachate treatment: A short review. *Journal of Environmental Chemical Engineering*, *10*(3), 107837.

[2] Vaverková, M. D., Zloch, J., Adamcová, D., Radziemska, M., Vyhnánek, T., Trojan, V., ... & Brtnický, M. (2019). Landfill leachate effects on germination and seedling growth of hemp cultivars (Cannabis Sativa L.). *Waste and Biomass Valorization*, 10, 369-376.

[3] Vaccari, M., Tudor, T. and Vinti, G. (2019), Characteristics of leachate from landfills and dumpsites in Asia, Africa and Latin America: an overview, *Waste Management*, 95, 416–431

[4] Boonnorat, J., Treesubsuntorn, C., Phattarapattamawong, S., Cherdchoosilapa, N., Seemuang-on, S., Somjit, M., ... & Prachanurak, P. (2021). Effect of leachate effluent water reuse on the phytotoxicity and micropollutants accumulation in crops. *Journal of Environmental Chemical Engineering*, 9(6), 106639.

[5] Cheremisinoff, N. P. (1997). Treating contaminated groundwater and leachate. *Groundwater remediation and treatment technologies*, 259-308.

[6] Chuangcham, U., Wirojanagud, W., Charusiri, P., Milne-Home, W., & Lertsirivorakul, R. (2008). Assessment of heavy metals from landfill leachate contaminated to soil: A case study of Kham Bon landfill, Khon Kaen province, NE Thailand. *Journal of Applied Sciences*, *8*(8), 1383-1394.

[7] Mojiri, A., Zhou, J. L., Ratnaweera, H., Ohashi, A., Ozaki, N., Kindaichi, T., & Asakura, H. (2021). Treatment of landfill leachate with different techniques: an overview. *Water Reuse*, *11*(1), 66-96.

[8] Ehrig, H., & Stegmann, R. (2019). Chapter 10.5-Combination of Different MSW Leachate Treatment Processes. *Solid Waste Landfilling: Concepts, Processes, Technologies, R. Cossu and R. Stegmann, Eds. Elsevier Inc*, 633-646.

[9] Renou, S., Givaudan, J. G., Poulain, S., Dirassouyan, F., & Moulin, P. J. J. O. H. M. (2008). Landfill leachate treatment: Review and opportunity. *Journal of hazardous materials*, *150*(3), 468-493.

[10] Wiszniowski, J., Robert, D., Surmacz-Gorska, J., Miksch, K., & Weber, J. V. (2006). Landfill leachate treatment methods: A review. Environmental chemistry letters, 4, 51-61.

[11] Chelliapan, S., Arumugam, N., Din, M. F. M., Kamyab, H., & Ebrahimi, S. S. (2020). Anaerobic treatment of municipal solid waste landfill leachate. In *Bioreactors* (pp. 175-193). Elsevier.

[12] Gautam, P., Kumar, S., & Lokhandwala, S. (2019). Advanced oxidation processes for treatment of leachate from hazardous waste landfill: A critical review. *Journal of Cleaner Production*, *237*, 117639.

[13] Kokkinos, P., Venieri, D., & Mantzavinos, D. (2021). Advanced oxidation processes for water and wastewater viral disinfection. A systematic review. *Food and Environmental Virology*, *13*(3), 283-302.

[14] Ma, D., Yi, H., Lai, C., Liu, X., Huo, X., An, Z., Li, L., Fu, Y., Li, B., Zhang, M., Qin, L., Liu, S., Yang. L. (2021). Critical review of advanced oxidation processes in organic wastewater treatment, *Chemosphere*, 275, 130104.

[15] Ran, G., & Li, Q. (2019). Removal of refractory organics in dinitrodiazophenol industrial wastewater by an ultraviolet-coupled Fenton process. *RSC advances*, *9*(44), 25414-25422.

[16] Bożym, M. (2020). Assessment of phytotoxicity of leachates from landfilled waste and dust from foundry. *Ecotoxicology*, *29*(4), 429-443.

[17] Rossi, R. (2019). Irrigation in EU agriculture.

[18] Halleux, V. (2020). Water reuse: Setting minimum requirements.

[19] Teng, C., Zhou, K., Peng, C., & Chen, W. (2021). Characterization and treatment of landfill leachate: A review. *Water research*, *203*, 117525.

[20] Parthenidis, P., Evgenidou, E., & Lambropoulou, D. (2023). Landfill leachate treatment by hydroxyl and sulfate radical-based advanced oxidation processes (AOPs). *Journal of Water Process Engineering*, *53*, 103768.

[21] Deng, Y., & Englehardt, J. D. (2006). Treatment of landfill leachate by the Fenton process. *Water Research*, *40*(20), 3683-3694.

[22] Klauck, C. R., Rodrigues, M. A. S., & Silva, L. B. (2015). Evaluation of phytotoxicity of municipal landfill leachate before and after biological treatment. *Brazilian Journal of Biology*, *75*, 57-62.

[23] Chin-kan, T. S. A. N. G. (2006). *Landfill Leachate Irrigation: Evaluation of Plant Productivity and Soil Toxicity* (Doctoral dissertation, The Chinese University of Hong Kong).

[24] Wong, M. H., & Leung, C. K. (1989). Landfill leachate as irrigation water for tree and vegetable crops. *Waste management & research*, *7*(4), 311-323.

Contribution of Individual Authors to the Creation of a Scientific Article (Ghostwriting Policy)

-Eleni Grilla and Ekavi Aikaterini Isari carried out the phytotoxicity experiments and wrote the manuscript draft.

-Periklis Parthenidis and Eleni Evgenidou did the leachate sampling, and characterization, and performed the experiments of Fenton and UV/Fenton treatment processes.

-Petros Kokkinos assessed the results and revised the manuscript.

-Ioannis Kalavrouziotis, and Dimitra Lambropoulou supervised the study and revised the manuscript.

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Conflict of Interest

The authors have no conflict of interest to declare.