Process Integration & Intensification

KEYWORDS: Multistage Treatment Approach / High-Low-Strength Wastewaters / Biological-Physical-Chemical Processes

Multistage treatment strategies, integrating both advanced and conventional treatment processes, were implemented to remediate high-strength and low-strength wastewaters, aiming their discharge into municipal sewage or water bodies or its reuse.

Objectives

Considering the variability and complex composition of municipal and industrial wastewater, relying on a singular treatment method might prove inadequate to achieve the quality levels imposed by current and future regulations, even more so when reuse is required. A treatment train strategy, integrating conventional and advanced treatment processes, enables the synergistic utilization of diverse technologies, effectively addressing the broad spectrum of pollutants present in wastewater. Therefore, the primary focus of this research project was the development of multistage treatment strategies for both (i) high-strength wastewater, encompassing landfill leachates, textile wastewater, and olive mill wastewater, and (ii) low-strength municipal wastewater.

Current Development

Four distinct sanitary landfill leachates originating from municipal and industrial solid waste landfills (MSWL and ISWL) were subjected to study: L1 – mature MSWL; L2 – a hazardous ISWL leachate; L3 – a non-hazardous leather tannery ISWL leachate; and L4 – a general non-hazardous ISWL leachate, in the framework of AIProcMat@N2020 project.

L1 leachate featured a high organic content with low biodegradability and high ammonium concentration. A full-scale facility was designed, constructed, and tested for treating 30 m^3 of leachate per day (Fig 1) in collaboration with EFACEC and SULDOURO companies.

Fig 1. Multistage treatment strategy for urban leachate remediation along with the main results (up) and scheme/photographs of the full-scale plant (bottom).

The multistage treatment system for L1 leachate consisted of: (i) biological oxidation to promote nitrification/denitrification reactions using glycerol as an external carbon source for total ammonium removal, and partial removal of alkalinity, nitrogen, and recalcitrant organics; (ii) coagulation/sedimentation (C/S), for partial removal of recalcitrant organics (mainly humic acids), color, and suspended solids; (iii) photo-Fenton (PF) reaction, for recalcitrant organics oxidation and biodegradability enhancement; and (vi) biological polishing step. Major operational difficulties were found in (i) the C/S stage due to the production of dense foam that trapped the sludge; and (ii) the PF stage for iron precipitation. These problems were overcome, respectively, with (i) sludge removal after nitrite oxidation with hydrogen peroxide (H_2O_2) (intermediate step before PF); and (ii) maintenance of a residual amount of H_2O_2 before the neutralization step. The sludge produced was also treated *in situ*, and compliance with legal disposal standards was verified.

L2 leachate was characterized by very high amounts of sulfur compounds and a high organic content with low biodegradability due to the presence of high amounts of sulfur compounds. The best treatment strategy comprised the following stages: (i) catalytic oxidation of sulfide and sulfite to sulfate using H_2O_2 as oxidant and metals present in the leachate as catalysts, (ii) chemical precipitation of sulfate as barite using barium chloride dehydrate, (iii) biological oxidation to degrade the biodegradable organics, (iv) coagulation/flocculation for partial removal of recalcitrant organics and suspended solids, (v) advanced oxidation process (AOP)/electrochemical AOP (EAOP)/ozonation for recalcitrant organics oxidation and biodegradability enhancement, and (vi) biological polishing step.

L3 leachate had a particularly high biodegradable organic content, extremely high levels of ammonium, and considerable levels of trivalent chromium. The best multistage treatment line for L3 leachate included: (i) continuous-flow anoxic/aerobic biological reactor comprising nitrification-denitrification for the removal of ammonium, alkalinity, and biodegradable organics; (ii) coagulation/flocculation for total removal of chromium and partial removal of recalcitrant organics, color and suspended solids; (iii) AOP/EAOP for recalcitrant organic matter oxidation and biodegradability improvement; and (iv) biological polishing step.

L4 leachate displayed moderate organic content with very low biodegradability and low ammonium concentration. Its remediation strategy included: (i) coagulation for partial removal of recalcitrant organics and color; (ii) AOP/EAOP/ozonation for recalcitrant organics oxidation and biodegradability enhancement; and (iii) final biological stage for removal of biodegradable organics and ammonia.

The treatment trains proposed provided final effluents in agreement with the 2013/39/EU European directive for the discharge into receiving water, except for the hazardous ISWL leachate, which complies with the limits imposed by urban wastewater treatment plants (WWTPs) for the discharge into the municipal sewage network. In general, ozonation was shown to be more versatile and easier to operate when compared to AOPs and EAOPs when applied in the oxidation of recalcitrant organics.

Moreover, a specific collaboration protocol was established with the SULDOURO sanitary landfill (LeachateCleanUp and LeachateBioCleaning projects) aiming at consulting and optimizing the leachate treatment plant, integrating biological and coagulation/flocculation steps up to levels that allow its discharge in the municipal collector, having in perspective discharge limit values to which the urban WWTP is subject.

The remediation of a real textile wastewater aiming for its reuse in the textile industry was carried out by integrating two processes: (i) a PF reaction for color removal and organics degradation; and (ii) a cation exchange process using the marine macroalgae (*Laminaria hyperborea*) for removal of the iron used in the PF reaction. The resulting textile wastewater was successfully reused in scouring, bleaching, and dyeing processes.

A treatment train for the remediation of raw olive mill wastewater was investigated, aiming to comply with the emission limit values for direct discharge into water bodies. The following stages were proposed: (i) pre-treatment by filtration and sedimentation for removal of oil/grease and solids/sludge, (ii) coagulation for removal of turbidity, solids, and phenols, decreasing the effluent turbidity, (iii) biological oxidation for degradation of organic matter; and (iv) advanced oxidation process (PF, ozonation and anodic oxidation) for organics oxidation.

Under the SERPIC project, an integral technology, based on a multi-barrier approach (Fig 2), to treat the effluents of municipal WWTPs was developed to maximize the reduction of contaminants of emerging concern (CECs). The project consortium includes 8 partners from 6 countries: Fraunhofer-Gesellschaft, Germany; SolarSpring GmbH, Germany; Universidad de Castilla-La Mancha, Spain; Università degli Studi di Ferrara, Italy; AdP VALOR, Serviços Ambientais, SA, Portugal; Faculty of Engineering University of Porto, Portugal; Norwegian Institute for Water Research, Norway; and Stellenbosch University, South Africa.

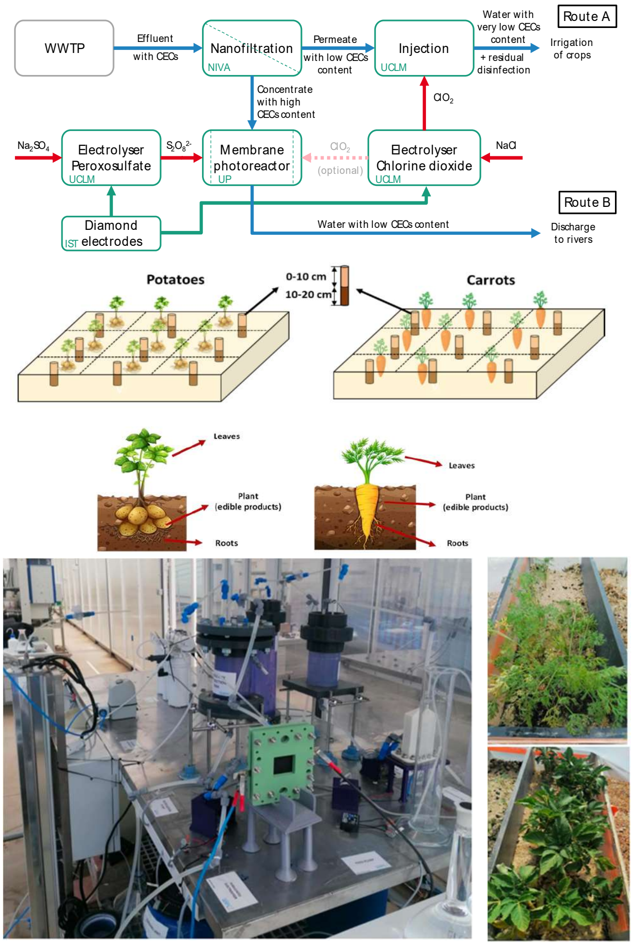

Fig 2. Scheme of the multi-barrier approach to treat the municipal WWTP effluent, and photographs of the prototype treatment plant and the agricultural tests.

Nanofiltration (NF) technology was applied to reduce CECs in the permeate stream by at least 90 % while retaining the nutrients. A residual disinfection using chlorine dioxide/ozone produced electrochemically was added to the stream used for crops irrigation (Route A). The CECs in the polluted concentrate (retentate) stream were reduced by at least 80 % by light-driven electro-chemical oxidation (UVC/persulfate) and ozonation. When discharged into the aquatic system (route B), it will contribute to the quality improvement of the surface water body. A prototype treatment plant was setup and evaluated for irrigation in long-term tests with the help of agricultural test pots (potatoes and carrots).

Future Perspectives

Future studies regarding MSWL leachates will focus on integrating aerobic granular sludge (AGS) technology with ozone-based processes.

Under the SERPIC project, transfer concepts will be developed to transfer the treatment technology results to other regions, especially in low- and middle-income countries.

Integration of ozonation, biofiltration, and membrane filtration for the quaternary treatment of urban wastewater, targeting its reuse for green hydrogen production by water electrolysis.

Related Sustainable Development Goals

Outputs

PhD Theses

[1] Alberto Dresch Webler, Tratamento de lixiviado de aterro industrial por processos combinados, UFRJ/FEUP, 2018
[2] Ana Isabel de Emílio Gomes, Treatment train for mature urban landfill leachate, PDEQB, FEUP, 2021
[3] Inalmar Dantas Barbosa Segundo, Multistage treatment system for leachates from industrial hazardous and non-hazardous waste landfills, PDEA, FEUP, 2021
[4] Sara Gabriela da Silva e Santos, A multistage treatment strategy for leachates from solid waste landfills: breaking barriers towards environmental compliance, PDEA, FEUP, on-going, 2020-2024

Selected Publications

[1] I.D. Barbosa et al., Sci. Total Environ. 655, 1246 (2019)
[2] A.I. Gomes et al., Chem. Eng. J., 376, 120573 (2019)
[3] A.D. Webler, Waste Manag. 89, 114 (2019)
[4] A.I. Gomes et al., Sci. Total Environ. 673, 470 (2019)
[5] I.D. Barbosa et al., Sci. Total Environ. 741, 140165 (2020)
[6] L.G. Silva, J. Environ. Manag. 272, 111082 (2020)
[7] I.D. Barbosa et al., J. Environ. Chem. Eng. 9, 105168 (2021)
[8] A.I. Gomes et al., Chemosphere 278, 130389 (2021)
[9] I.D. Barbosa et al., J. Environ. Chem. Eng. 9, 105554 (2021)
[9] S. Vuppala et al., J. Environ. Chem. Eng. 10, 107442 (2022)

Team

Vítor Vilar Principal Researcher; Rui Boaventura Principal Researcher; Ramiro J. Professor; Ricardo J. Auxiliar Researcher; Tânia Silva Junior Researcher; Francisca Moreira Junior Researcher; **Ana Isabel Gomes** Junior Researcher; **Alberto Webler**, PhD Student; **Inalmar Segundo**, PhD Student; **Laís Silva**, PhD Student; **Srikanth Vuppala**, PhD Student; **Daniela Morais**, PhD student; **Sara Santos**, PhD student; **Carla Santos**, PhD student; **Mateus Caixeta**, PhD student; **Augustina Olivera**, PhD student

Funding

AlProcMat@N2020, NORTE-01-0145-FEDER-000006, 2016-2019
LeachateCleanUp, FEUP/SULDOURO/2019-74570, 2019-2020
LeachateBioCleaning, FEUP/SULDOURO/2022-79814, 2022-2023
SERPIC, Aquatic/0002/2020, 2021-2024
BagaÇo+Valor, NORTE-01-0247-FEDER-072124, 2020-2023
LSRE-LCM Base Funding, UIDB/50020/2020, 2020-2023
LSRE-LCM Programmatic Funding, UIDP/50020/2020, 2020-2023
LA LSRE-LCM Funding, UID/EQU/50020/2019, 2019
LA LSRE-LCM Funding, POCI-01-0145-FEDER-006984,2013-2018
FCT Grants: CEECIND/01317/2017, CEECIND/01386/2017, CEECIND/02196/2017
FCT Scholarships: PD/BD/105980/2014, SFRH/BD/146476/2019, 2020.04970. BD, 2021.07416.BD, 2022.10796.BD, 2020.06681.BD
Other Scholarships: CNPq/202372/2015-4, CNPq/ 205715/2014-1