

Wastewater Treatment, Valorization, and Reuse

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This Special Issue includes investigations related to wastewater treatment, recovery, and reuse. Different techniques including adsorption, electrocoagulation, ultrafiltration, and membrane filtration were applied to eliminate heavy metals and organic compounds. Furthermore, the use of green technique for water, nutrients and polyhydroxyalkanoates recovery from wastewater was tested. Finally, the potential of the treated wastewater reuse in region under hydric stress was assessed. The different published papers show the diversity of research conducted on wastewater treatment and the environmental and economic benefits from their reuse in various applications. New developments are still in progress, encouraging the organization of another edition of this Special Issue.

Mineral processing wastewater contains large amounts of reagents which can lead to severe environmental problems, such as high chemical oxygen demand (COD). Inspired by the wastewater treatment in such industries as those of textiles, food, and petrochemistry, Jing et al. [1] have applied electrocoagulation (EC) for the first time to explore its feasibility in the treatment of wastewater with an initial COD of 424.29 mg/L from a Pb/Zn sulfide mineral flotation plant and its effect on water reuse. Typical parameters, such as anode materials, current density, initial pH, and additives, were characterized to evaluate the performance of the EC method. The results showed that, under optimal conditions, i.e., iron anode, pH of 7.1, electrolysis time of 70 min, 19.23 mA/cm² of current density, and 4.1 g/L of activated carbon dose, the initial COD can be reduced to 72.9 mg/L, corresponding to a removal rate of 82.8%. In addition, compared with the untreated wastewater, EC-treated wastewater was found to benefit the recovery of galena and sphalerite, with galena recovery increasing from 25.01% to 36.06% and sphalerite recovery increasing from 59.99% to 65.33%. This study confirmed that EC is a promising method for the treatment and reuse of high-COD-containing wastewater in the mining industry, and it possesses great potential for a wide range of industrial applications.

Othmani et al. [2] have synthesized hybrid materials with high pollutant-uptake capacity and low cost based on *Luffa cylindrica* (L.C) and different percentage of Zn²⁺ in the presence and absence of alternating current (AC). Physico-chemical, morphological, and structural characterizations of the hybrid materials were performed by Boehm method, point zero charge (pH_{pzc}), infrared characterizations (IR), scanning electron microscopy (SEM), energy-dispersive spectroscopy, and X-ray photoelectron spectroscopy. The efficiency of the designed hybrid materials was optimized based on their performance in water depollution. Methylene blue (MB) and industrial textile wastewater were the investigated pollutants models. IR characterizations confirmed the fixation of Zn²⁺ onto the L.C by the creation of Zn–OH, Zn–O and Zn–O–C bonds. Boehm titration showed that the fixation of Zn²⁺ onto L.C is accompanied by an increase in the basic functions of its surface, and subsequently an increase in the pH_{pzc}. SEM results confirmed the fixation of Zn²⁺ onto the L.C. Coupling AC with biosorption showed an increase in the adsorbed amount of MB and speed. When adding 4% of Zn²⁺ compared to the pure L.C, the Q_m shifted from 3.22 to



Citation: Jeguirim, M.; Jellali, S. Wastewater Treatment, Valorization, and Reuse. *Water* **2021**, *13*, 548. <https://doi.org/10.3390/w13040548>

Received: 16 January 2021

Accepted: 18 February 2021

Published: 21 February 2021

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9.84 and 8.81 mg/g for hybrid materials synthesized under AC, in an absence of AC, and pure *L.C.*, respectively.

Miron et al. [3] have studied the evolution of the membrane selectivity of neutral solutes after the filtration of protein or amino acid solutions. Classical methodologies led to the estimation of the mean pore radius, different for each filtrated neutral solute. The use of pore size distribution from nitrogen adsorption/desorption experiments enabled a good description of hydraulic and selectivity performances. The modification of the membrane hydraulic properties after the successive filtration of protein solutions revealed that the decrease is quasi linear, the same for all the studied membranes, and independent of prior tests. According to the experimental observations, an adsorption model was developed, considering a layer-by-layer adsorption in the larger pores of the membrane. The predictive obtained results are in good agreement with the experimental rejection rates, validating the assumptions.

Lignite, as an available and low-cost material, was tested for cadmium (Cd) and copper (Cu) removal from aqueous solutions under various static experimental conditions by Jellali et al. [4]. Experimental results showed that the removal efficiency of both metals was improved by increasing their initial concentrations, adsorbent dosage, and aqueous pH values. The adsorption kinetic was very rapid for Cd; about 78% of the totally adsorbed amounts were removed after a contact time of only 1 min. For Cd and Cu, the kinetic and isothermal data were well fitted with pseudo-second order and Freundlich models, respectively, which suggests that Cd/Cu removal by lignite occurs heterogeneously on multilayer surfaces. The maximum Langmuir's adsorption capacities of Cd and Cu were assessed to 38.0 and 21.4 mg g⁻¹ and are relatively important compared to some other lignites and raw natural materials. Results of proximate, scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS), Fourier-transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD) showed that the removal of these metals occurs most likely through a combination of cation exchange and complexation with specific functional groups. The relatively high adsorption capacity of the used lignite promotes its future use as a low-cost material for Cd and Cu removal from effluents, and possibly for other heavy metals or groups of pollutants.

Greywater has been identified as a potential source of water in a number of applications, e.g., toilet flushing, laundering in first rinsing, floor cleaning, and irrigation. The major obstacle to the reuse of greywater relates to its relatively high contents of pathogens, nutrients, and organic matter. Therefore, much effort has been paid to treat greywater, in order to yield high-quality water deprived of bacteria and with an appropriate value in a wide range of quality parameters (total organic carbon (TOC), nitrate, phosphate, ammonium, pH, and absorbance), similar to the values for tap water. Kamińska and Marszałek [5] proposed to treat real greywater and turn it into high-quality and safe water. For this purpose, the real greywater was treated by means of a sequential biological reactor (SBR) followed by ultrafiltration. Initially, greywater was treated in a laboratory SBR reactor with a capacity of 3 L, operated in a 24 h cycle. Then, SBR effluent was purified in a cross-flow ultrafiltration setup. Treatment efficiency in SBR and ultrafiltration was assessed using extended physicochemical and microbiological analyses (pH, conductivity, color, absorbance, chemical oxygen demand (COD), biological oxygen demand (BOD₅), nitrate, phosphate, ammonium, total nitrogen, phenol index, nonionic and anionic surfactants, TOC, *Escherichia coli*, and enterococci). Additionally, ultrafiltration was evaluated in terms of fouling behavior for three polymer membranes with different MWCOs (molecular weight cut-offs). The values of quality parameters (pH, conductivity, COD, BOD₅, TOC, N-NH₄⁺, N-NO₃⁻, N_{tot}, and P-PO₄³⁻) measured in SBR effluent did not exceed permissible values for wastewater discharged to soil and water. Ultrafiltration provided the high-quality water with very low values of COD (5.8–18.1 mg/L), TOC (0.47–2.19 mg/L), absorbance UV254 (0.015–0.048 1/cm), color (10–29 mgPt/L), and concentration of nitrates (0.18–0.56 mg/L), phosphates (0.9–2.1 mg/L), ammonium (0.03–0.11 mg/L), and total nitrogen (3.3–4.7 mg/L), as well as

a lack of *E. coli* and enterococci. Membrane structural and surface properties did not affect the treatment efficiency, but did influence the fouling behavior.

Water shortage is a very concerning issue in the Mediterranean region, threatening the viability of the agriculture sector, and in some countries, population wellbeing. At the same time, liquid effluent volumes generated from agro-food industries in general, and the olive oil industry in particular, are huge. Thus, Dutournié et al. [6] proposed a sustainable solution for the management of olive mill wastewater (OMWW) with possible reuse in irrigation. Their investigation is a part of a series of papers valorizing all the outputs of a three-phase system of olive oil mills. It deals with recovery, by condensation, of water from both OMWW and OMWW-impregnated biomasses (sawdust and wood chips), during a convective drying operation (air velocity: 1 m/s and air temperature: 50 °C). The experimental results showed that the water yield recovery reaches about 95%. The condensate waters have low electrical conductivity and salinities, but also acidic pH values and slightly high chemical oxygen demand (COD) values. However, they could be returned suitable for reuse in agriculture after additional low-cost treatments.

Raw poultry manure (RPM) and its derived biochars at temperatures of 400 (B400) and 600 °C (B600) were physico-chemically characterized, and their ability to release nutrients was assessed under static conditions by Hadroug et al. [7]. The experimental results showed that RPM pyrolysis operation significantly affects its morphology, surface charges, and area, as well as its functional groups contents, which in turn influences its nutrient release ability. The batch experiments indicated that nutrient release from the RPM as well as biochars attains a pseudo-equilibrium state after a contact time of about 48 h. RPM pyrolysis increased phosphorus stability in residual biochars and, in contrast, transformed potassium to a more leachable form. For instance, at this contact time, P- and K-released amounts changed from 5.1 and 25.6 mg g⁻¹ for RPM to only 3.8 and more than 43.3 mg g⁻¹ for B400, respectively. On the other hand, six successive leaching batch experiments with a duration of 48 h each showed that P and K release from the produced biochars was a very slow process; negligible amounts continued to be released even after a total duration of 12 days. All these results suggest that RPM-derived biochars have specific physico-chemical characteristics, allowing them to be used in agriculture as low-cost and slow-release fertilizers.

Sludge from municipal wastewater treatment systems can be used as a source of mixed microbial cultures for the production of polyhydroxyalkanoates (PHA), according to de Souza Reis et al. [8]. Stored intracellularly, the PHA is accumulated by some species of bacteria as an energy stockpile and can be extracted from the cells by reflux extraction. Dimethyl carbonate was tested as a solvent for the PHA extraction at different extraction times and biomass-to-solvent ratios, and 1-butanol was tested for purifying the obtained PHA at different purification times and PHA-to-solvent ratios. Overall, only a very small difference was observed in the different extraction scenarios. An average extraction amount of 30.7 ± 1.6 g of PHA per 100 g of biomass was achieved. After purification with 1-butanol, a visual difference was observed in the PHA between the tested scenarios, although the actual purity of the resulting samples did not present a significant difference. The overall purity increased from $91.2 \pm 0.1\%$ to $98.0 \pm 0.1\%$.

Current regulations and legislation require critical revision to determine safety for alternative water sources and water reuse as part of the solution to the global water crisis. In order to fulfill those demands, the Lisbon municipality decided to start water reuse as part of a sustainable hydric resource management, and there was a need to confirm safety and safeguard for public health for its use in this context. For this purpose, a study was designed that included a total of 88 samples collected from drinking, superficial, underground water, and wastewater at three different treatment stages [9]. Quantitative polymerase chain reaction (PCR) detection (qPCR) of enteric viruses norovirus (NoV) genogroups I (GI) and II (GII) and hepatitis A (HepA) was performed, and FIB (*E. coli*, enterococci and fecal coliforms) concentrations were also assessed. HepA virus was only detected in one untreated influent sample, whereas NoV GI/NoV GI were detected in

untreated wastewater (100/100%), secondary treated effluent (47/73%), and tertiary treated effluent (33/20%). This study proposes that NoV GI and GII should be further studied to provide the support that they may be suitable indicators for water quality monitoring targeting wastewater treatment efficiency, regardless of the level of treatment.

Water scarcity remains the major looming challenge which is facing Jordan. Wastewater reclamation is considered as an alternative source of fresh water in semi-arid areas with water shortage or increased consumption. The current status of wastewater reclamation and reuse in Jordan was analyzed considering 30 wastewater treatment plants (WWTPs) [10]. The assessment was based on the WWTPs' treatment processes in Jordan, the flowrate scale, and the effluents' average total dissolved solid (TDS) contents. Accordingly, 60% of the WWTPs in Jordan used activated sludge as a treatment technology; 30 WWTPs were small scale ($<1 \times 10^4$ m³/day); and a total of 17.932 million m³ treated wastewater had low TDS (<1000 ppm) that generally can be used in industries with relatively minimal cost of treatment. Moreover, the analysis classified the 26 million m³ groundwater abstraction by major industries in Jordanian governorates. The results showed that the reclaimed wastewater can fully offset the industrial demand of fresh water in the Amman, Zarqa, and Aqaba governorates. Hence, the environmental assessment showed positive impacts of reclaimed wastewater reuse scenario in terms of water depletion (saving of 72.55 million m³ groundwater per year) and climate change (17.683 million kg CO_{2Eq} reduction). The energy recovery assessment in the small- and medium-scale WWTPs ($<10 \times 10^4$ m³/day) revealed that generation of electricity by anaerobic sludge digestion equates potentially to an offset of 0.11–0.53 kWh/m³. Finally, several barriers and prospects were imposed to help the stakeholders when considering entering into an agreement to supply and/or reuse reclaimed water.

We found the edition and selections of papers for this Special Issue very inspiring and rewarding. We also thank the editorial staff and reviewers for their efforts and help during the process.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Jing, G.; Ren, S.; Gao, Y.; Sun, W.; Gao, Z. Electrocoagulation: A Promising Method to Treat and Reuse Mineral Processing Wastewater with High COD. *Water* **2020**, *12*, 595. [\[CrossRef\]](#)
2. Othmani, A.; Kesraoui, A.; Boada, R.; Seffen, M.; Valiente, M. Textile Wastewater Purification Using an Elaborated Biosorbent Hybrid Material (*Luffa-Cylindrica*–Zinc Oxide) Assisted by Alternating Current. *Water* **2019**, *11*, 1326. [\[CrossRef\]](#)
3. Miron, S.M.; Dutournié, P.; Ponche, A. Filtration of Uncharged Solutes: An Assessment of Steric Effect by Transport and Adsorption Modelling. *Water* **2019**, *11*, 2173. [\[CrossRef\]](#)
4. Jellali, S.; Azzaz, A.A.; Jeguirim, M.; Hamdi, H.; Mlayah, A. Use of Lignite as a Low-Cost Material for Cadmium and Copper Removal from Aqueous Solutions: Assessment of Adsorption Characteristics and Exploration of Involved Mechanisms. *Water* **2021**, *13*, 164. [\[CrossRef\]](#)
5. Kamińska, G.; Marszałek, A. Advanced Treatment of Real Grey Water by SBR Followed by Ultrafiltration—Performance and Fouling Behavior. *Water* **2020**, *12*, 154. [\[CrossRef\]](#)
6. Dutournié, P.; Jeguirim, M.; Khiari, B.; Goddard, M.-L.; Jellali, S. Olive Mill Wastewater: From a Pollutant to Green Fuels, Agricultural Water Source, and Bio-Fertilizer. Part 2: Water Recovery. *Water* **2019**, *11*, 768. [\[CrossRef\]](#)
7. Hadroug, S.; Jellali, S.; Leahy, J.J.; Kwapinska, M.; Jeguirim, M.; Hamdi, H.; Kwapinski, W. Pyrolysis Process as a Sustainable Management Option of Poultry Manure: Characterization of the Derived Biochars and Assessment of their Nutrient Release Capacities. *Water* **2019**, *11*, 2271. [\[CrossRef\]](#)
8. De Souza Reis, G.A.; Michels, M.H.A.; Fajardo, G.L.; Lamot, I.; de Best, J.H. Optimization of Green Extraction and Purification of PHA Produced by Mixed Microbial Cultures from Sludge. *Water* **2020**, *12*, 1185. [\[CrossRef\]](#)
9. Teixeira, P.; Costa, S.; Brown, B.; Silva, S.; Rodrigues, R.; Valério, E. Quantitative PCR Detection of Enteric Viruses in Wastewater and Environmental Water Sources by the Lisbon Municipality: A Case Study. *Water* **2020**, *12*, 544. [\[CrossRef\]](#)
10. Saidan, M.N.; Al-Addous, M.; Al-Weshah, R.A.; Obada, I.; Alkasrawi, M.; Barbana, N. Wastewater Reclamation in Major Jordanian Industries: A Viable Component of a Circular Economy. *Water* **2020**, *12*, 1276. [\[CrossRef\]](#)