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An innovative solution for managing waterworks sludge: Developing an alum sludge-based multi-stage constructed wetland system for wastewater treatment

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Abstract

Waterworks sludge continues to be an inescapable by-product of the potable water treatment process. Accordingly, final disposal of the sludge remains one of the most significant pressing problems for the potable water treatment industry. The possibility of reusing the sludge as a main substrate in a novel constructed wetland system was investigated in this study. Results show that significant phosphorus (P) and other pollutants removal were achieved in the system. With a mean influent \( \text{BOD}_5 \) (5-day biochemical oxygen demand) and COD (chemical oxygen demand) levels of 392.7 mg/l and 579.8 mg/l, respectively, a removal efficiency of 90.6 % and 71.8 %, respectively, was obtained. P removal was however exceptionally high despite the high influent mean P level of 45.3 mg-P/l, which is about 2-3 times the level of P commonly found in sewage. This is attributable to the P adsorption capacity of the alum sludge and this highlights the benefits of its reuse in the system. The paper presents and discusses the findings from a laboratory scale research, which has potential for further large scale implementation.
**Key words:** aluminium, alum sludge, constructed wetland, phosphorus, reuse, wastewater treatment

**Introduction**

Alum sludge refers to the residual by-product obtained as a result of the potable water treatment processes including coagulation (using aluminium salts), flocculation, clarification and filtration. It is the most widely generated waterworks sludge worldwide. Upon the sludge treatment processes (thickening, conditioning and dewatering) a residual solid cake referred to as dewatered alum sludge cake (DASC) is obtained. In Ireland and most countries worldwide, the DASC is treated as a waste. In particular, the official position in Ireland is that it is largely inert in nature and has little value in reuse applications. Based on this, they are totally consigned to landfills. However, giving the inevitability of DASC generation and the escalating costs and concerns associated with its disposal in landfills, a forced re-think of the current approach of regarding DASC as a waste and landfilling it is inevitable. Globally, there are now about eleven different ways in which waterworks sludges are being reused or recycled, but none has entailed its use as a resourceful material for wastewater treatment [1].

At the University College Dublin (UCD), Ireland, a new and innovative reuse option for DASC is currently being investigated and it involves the reuse of DASC as main substrate in a novel constructed wetland system (CWs). CWs have been used successfully worldwide as one of the most popular technologies to treat various types of wastewaters [2-5]. They are considered to be efficient and at the same time economically and environmentally attractive and sustainable. In the last several years, extensive studies have been carried out at UCD to develop novel approaches for the purpose of enhancing wastewater treatment and pollutant removal in such CWs.
One of such studies has focused on the innovative reuse of DASC in a tidal-flow CWs operated in the multi-stage format, for the purpose of enhancing the removal of P and organic matter. The use of DASC as main substrate in such CWs is based on the fact that the abundant aluminium (Al) ions in the DASC gives it a very strong affinity for P immobilization, thereby providing an opportunity to reuse the DASC as a resourceful adsorbent for P removal and enhanced wastewater treatment in CWs. This presents a 2-in-1 strategy of managing waterworks sludge and wastewater treatment using the novel multi-stage CWs. P present in wastewater is of significant environmental importance because it is an essential, often limiting nutrient for plants and microorganisms. Therefore, if it is not effectively removed, its release to receiving water causes eutrophication and has negative impact on the ecosystem. Apart from P, organic constituents in wastewater (both soluble and insoluble forms) are oxygen depleting and are associated with unpleasant color and odour, growth substrates for bacteria and can even cause health problems. These are some reasons why removal of P and organics from wastewater is important before discharge to sensitive water bodies.

Although P removal in CWs can occur through a combination of several processes, adsorption and precipitation within the substrate are widely acknowledged and known to play the greatest role. Therefore, to ensure efficient P removal, it is important to use substrates with high P removal capacity and suitable physicochemical properties. The tidal flow operation strategy for the CWs is a batch wise, fill-and-draw type operation [6,7]. When a periodic influent feeding and periodic discharge is applied, it enables the bed matrices to be fully submerged during the filling process. This provides maximum media-wastewater contact and avoids the problem of poor wastewater distribution often associated
with conventional continuous-flow CWs. Subsequent draining process then allows air to be drawn from the atmosphere into the bed matrices without mechanical input, thereby enhancing aeration and stimulating aerobic biological processes to decompose organic pollutants and ammoniacal-nitrogen in wastewater. This paper presents and discusses the results obtained from a laboratory trial of the novel tidal flow CWs using DASC as the main substrate.

**Materials and Methods**

**Dewatered alum sludge cakes**

DASC were obtained from the mechanical dewatering unit of the Ballymore-Eustace Water Treatment Works, South-west, Dublin, Ireland. The plant uses aluminium sulphate as primary coagulant for reservoir water flocculation and it serves the water needs of approximately one-third of the entire Irish population, producing 230,000 m$^3$/d of high quality potable water. About 45-75 tonnes of DASC are generated at this plant each day, and this is entirely sent to landfills at a cost of about €130/tonne. Upon collection, the DASC was air-dried and ground and then used as the main substrate in a novel CWs. The ground DASC had a $d_{10}$, $d_{60}$ and uniformity coefficient of 0.5 mm, 1.8 mm and 3.6 mm respectively [8], which are consistent with several national guidelines for CWs substrates [9].

**The constructed wetland system and operation**

The lab-scale CWs consist of 4 stages of individual constructed wetland units which are all linked together using peristaltic pumps (Fig 1). The individual constructed wetland units are all identical and are constructed using Pyrex tubes (1.0 m long, $\varnothing = 0.095$ m).
Each of the unit had 10 cm of 6-10 mm gravel at the base to serve as support. Prepared alum sludge was filled into the wetland as main substrate up to a depth of 50 cm. Young *Phragmites australis* obtained from a local supplier was planted on top of each bed. The lab-scale CWs was operated using the tidal flow strategy which involves rhythmically filling and draining the individual constructed wetland matrices with wastewater, as the wastewater passes sequentially from the first to the last unit. The rhythmical filling and draining generated the tides, and this was realised by peristaltic pumps controlled by a preset electronic timer. The rhythmical operation was carried out in cycles. Each cycle consist of 1 hour of wastewater contact with the alum sludge in the bed and 3 hours of resting, during which the wastewater is drained out from the bed and the system is allowed to rest before the next cycle commences. A hydraulic loading rate of 1.27 m$^3$/m$^2$.d was used.

The wastewater used was collected from the secondary holding tank of an animal research farm and it had a concentration of 322-510 mg/l (SS), 720-1523 mg/l (COD, chemical oxygen demand), 540-850 mg/l (BOD$_5$, 5-day biochemical oxygen demand), 48-73 mg/l (P) and 6.7-7.4 (pH). On being brought to the laboratory, the wastewater was allowed to settle overnight and the supernatant was collected the following morning. Depending on the concentration of pollutants in the supernatant, dilution was carried out before it is fed in to the lab-scale system. Accordingly, the influent wastewater into the system had mean concentrations of 392.7 mg/l (BOD$_5$), 579.8 (COD), 21.0 mg/l (SRP, soluble reactive phosphorus) and 45.3 mg/l (RP, reactive phosphorus).
Sampling and measurements

At specific periods during each operational week, wastewater samples were collected from the feed tank, and from the inlet and outlet of each stage of the system and analyzed for BOD$_5$, COD and P-PO$_4^{3-}$ (including both SRP determined on filtered samples, and RP determined on unfiltered samples). BOD$_5$ and COD were analysed using the dilution method and the closed reflux titrimetric method respectively according to sections 5210-B and 5220-C of the Standard Examination of Water and Wastewater [10]. All P analyses were done using a Hach DR/2400 spectrophotometer. Furthermore, specific metal levels were monitored in both the influent and the effluent on three occasions, while the increase or decrease in the dissolved elemental metal concentration of the DASC was also assessed to monitor any release of substances from the DASC into the effluent. To obtain the total dissolvable metals in the DASC, an Anton Paar MULTIWAVE microwave sample preparation system was used to digest the samples. Approximately 0.025 g of the DASC samples was weighed into clean TMF (trifluoromethylene) vessels followed by the addition of 4 ml HNO$_3$ + 200 µl HF + 4 ml H$_2$O. A built-in computer program was then used to specify the decomposition program, control the MULTIWAVE, and hold a library of sample data. When decomposition is complete, the sample is transferred to a volumetric flask and the volume is made up to 15 ml. These were then sent out to a certified laboratory for dissolved metal analysis using the ICP-MS (Inductively Coupled Plasma - Mass Spectrometer). Total and dissolved metal analysis on the influent and effluent samples was also done through the contracted certified laboratory using ICP (IRIS) and ICP-MS for the total and dissolved metals respectively.
Results and discussion

The treatment performance obtained in the DASC based CWs is presented in Fig. 2. From Fig. 2 (top), it can be seen that a significant reduction of both SRP and RP was obtained and this demonstrates one of the primary goals of the study, which is to show that the DASC can be beneficially reused to enhance P removal in CWs. By reusing the DASC as a substrate in the novel CWs, the need and the amount of DASC for landfilling can be reduced while at the same time, significant performance improvements and reduction in overall capital cost of CWs can be achieved. This presents a 2-in-1 approach to managing waterworks sludge and enhancing wastewater treatment in CWs. Average removal percentages of 97.6% (SRP) and 93.3% (RP) were achieved in the system at influent P concentrations of 21.0 mg/l (SRP) and 45.3 mg/l (RP). The P removal results are far better than the average results obtained on P removal in CWs and this is attributable to the strong affinity for P adsorption by the DASC, based on the fact that the DASC can significantly adsorb P from aqueous solution through complexation and ligand exchange mechanism [11]. P removal in most constructed wetlands/reed beds is often varied and mostly poor. One of the major reasons for the poor P removal is the poor P adsorption capacity of the media often used in such wetlands which are usually gravels and/or local soils. It should be noted that irrespective of the type of CWs, adsorption and/or precipitation on the substrate material used as media in the CWs is believed to play the major role in P removal from CWs and the only option open to engineering modification [12]. Consequently, several materials have been tested as potential adsorbents to provide enhanced sorption capacity towards improving P removal in CWs [13]. Braskerud [14] reported that surface flow CWs in Norway retained 21-44 % of Total-P non-source inputs from agricultural drainage waters, most of which were in particulate form,
while in Connecticut, USA, a farm scale CWs used to treat point source agricultural waters retained up to 68% of the incoming TP from a milk house dairy unit [15].

[INSERT FIG 2 HERE]

In Ireland, Healy and Cawley [16] reported an average removal of 13% (Total-P) and 26% (Ortho-P) for a tertiary CWs while O’Hogain [17] reported an overall average phosphate removal of 26% for a hybrid system treating municipal wastewater. The hybrid system had a 0% percentile compliance with EU discharge guidelines for P. In North America, a database comprising performance data from over a hundred CWs and compiled for the US-EPA by Knight et al., [18] shows that P removal was mostly lower, compared to other pollutants, and it is generally subject to saturation after prolonged loading. However, notwithstanding the fact that the influent P concentration used in this study is about 2-3 times the average level in typical domestic sewage, significant P removal was still achieved, thus demonstrating the beneficial effect of DASC inclusion in the system and offering an innovative way of reusing the DASC as opposed to its landfilling. This is where the DASC based CWs being demonstrated in this paper has a clear advantage. The alum sludge has a very high adsorption capacity compared to the gravel and local soils. In addition, although there are other by-products and materials that have been proposed for enhancing P removal in CWs, alum sludge still has another huge advantage in that it is easily, largely, locally and freely available in almost everywhere in the world. In our previous batch-isotherm studies, the P adsorption capacity of the DASC was determined to range from 10.2 to 31.9 mg-P/g (using the Langmiur adsorption isotherm) and the adsorption capacity of the DASC was
found to be in the high end range of capacities reported in the literature for several materials [12]. In Ireland where surface waters are at an increasing risk of being eutrophied as a result of high level of P in run offs and discharges, reusing the DASC either in the novel DASC based CWs or any other form of CWs can be integrated as part of a management strategy to intercept and reduce P in the runoffs and discharges. Regarding how long the system can continue to remove P, it has been shown that by considering the adsorption proportion in overall P removal, the lifetime of the DASC can be 9-40 years [19]. Even in the case of high P containing wastewater, the lifetime of DASC can be estimated as 2.5-3.7 years. However, for multi-stage systems, the lifetime is reasonably expected to be longer. Furthermore, upon saturation of the DASC with P, it has potential for P recovery [20].

In addition to the enhanced P removal efficiency of the system, significant removals of both BOD₅ and COD were also achieved in the system which again, illustrates the development of biofilm coupled with intensive biological activities inside the system. Fig. 2 (bottom) shows the performance of the system for the removal of BOD₅ and COD. It can be seen that average removal efficiencies of 90.6 % for BOD₅ and 71.8 % for COD were achieved in the system. In typical CWs where the reed plants are assumed to transfer a significant portion of oxygen needed for aerobic microbial degradation into the system, this may have not been possible. The significant removal efficiency obtained in the CWs can be attributed to the tidal flow strategy of operation of the system which has an oxygen transfer potential of about 0.378 g-O₂ for each individual stage of the CWs per each cycle [12]. From Fig. 2 (bottom), the $C_{BOD}/C_{COD}$ of the influent wastewater ranged between 0.592–0.915, which indicates very high biodegradability. However, the average $C_{BOD}/C_{COD}$ in the effluent exiting the system was 0.23 and this implies that the biodegradability of the effluent exiting
the system is quite low. This shows that a major proportion of the organic load in the effluent was biodegraded and this can be adduced to the enhanced oxygen transfer efficiency of the tidal flow strategy. It is expected that as the system matures further, more significant BOD₃ and COD removal can be achieved.

In order to examine the possibility of the leaching of metal constituents in the DASC into the treated wastewater, levels of metals in the sludge before and after use and also in the influent and effluent were monitored and some of the results are presented in Table 1. Aluminium was chosen being that the primary coagulant used in the treatment plant where the DASC was obtained is an aluminium salt. Other metals tested were selected based on their relative presence in the DASC according to the initial analysis for the DASC obtained from the water treatment plant operators. Table 1 shows that there were reductions in the levels of Al, arsenic (As), Fe, lead (Pb) and manganese (Mn) in the used DASC whereas the concentrations of P, calcium (Ca), magnesium (Mg), zinc (Zn) and titanium (Ti) were increased. From the table, it can be seen that the level of Al (both total and dissolved) in the effluent was higher than that in the influent (feed) in all the periods. This suggests some release of Al into the treated wastewater during passage through the DASC based CWs. However, based on the level of total and dissolved concentration of Al in the influent (feed) as shown in the table, it can be inferred that most of the Al in the influent is probably associated with the solids. Ca in the influent wastewater was mostly in the soluble form, probably associated with CaCO₃.

[INSERT TABLE 1 HERE]
However, there was a huge reduction in the level of Ca in the effluent exiting the system as compared to the Ca levels in the influent and this is likely due to the adsorption/precipitation of Ca ions onto the DASC. The concentration of Pb in the effluent (both total and dissolved) ranged between 0.001 to 0.05 mg/l and this is within and/or about the prescribed limit of 0.05 mg/l for drinking waters [21]. Mg and Zn levels were decreased across some periods, but their levels were increased in the used DASC and this shows the ability of the DASC to further adsorb Mg and Zn from the wastewater. A detailed analysis of these metal levels and their comparison with respective environmental quality standards (EQS) has been carried out in our other studies [12]. Our main findings have shown that dissolved levels of lead and arsenic in the effluents range from <1 μg/l to 5 μg/l and 3μg/l to 63 μg/l respectively, and these were below the prescribed limits of 50μg/l (except arsenic on one instance) for their respective discharge into freshwaters based on comparison with EQS [22]. However, dissolved aluminium ranged from 58 μg/l to 1106 μg/l with about 70 % of samples above prescribed limit of 200 μg/l for aluminium. Notwithstanding, the effluent is still shown to have some reuse potential in agriculture. It should however be pointed out that the possible release of polymer residual to the effluent from DASC still needs to be investigated since polymer is normally added as sludge conditioner to enhance its dewaterability. In particular, polymer residual in the DASC and its potential release when the sludge is reused needs to be studied since the toxicity of degraded polymer in the environment remains an unknown health risk from the long term point of view [23]. Overall, this attempt has demonstrated an ample opportunity for the beneficial reuse of DASC as a raw material in wastewater treatment engineering, thereby opening up an innovative reuse alternative.
Conclusions

This study has shown the potential use of dewatered alum sludge cakes (DASC) as a constructed wetland substrate, thereby transferring the dewatered alum sludge cakes from waste into a useful material. Results from the lab scale study shows that very high pollutant removal efficiencies can be obtained, thereby justifying the proposition that constructed wetland systems constructed using the sludge cakes as the main substrate can be operated successfully to obtain high pollutant removal efficiencies. Over the study period, average removal efficiencies of COD (90 %), BOD$_5$ (88 %), PO$_4^{3-}$-P (91 %), TN (76 %), SS (91 %) was achieved and this presents a superior performance when compared to traditional constructed wetland systems. A major conclusion of this study is that, instead of continuous landfilling of the DASC at huge costs, the DASC can be reused as a main constructed wetland substrate to significantly reduce P and other pollutants concentration in wastewaters, and thereby, contribute to reducing eutrophication of surface waters and improving water quality. However, there were release of certain metals from the DASC into the effluent and as such, further studies, including examination of possible polymer release from the DASC are necessary.

Acknowledgements

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References


Table 1. Input/output data for examining elemental metal leaching in the constructed wetland system

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<tr>
<th>Parameter</th>
<th>Al</th>
<th>As</th>
<th>Ca</th>
<th>Fe</th>
<th>Pb</th>
<th>Mg</th>
<th>Mn</th>
<th>P</th>
<th>Ti</th>
<th>Zn</th>
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<td>unused DASC (mg/g)</td>
<td>42.696</td>
<td>0.0336</td>
<td>0.8196</td>
<td>3.258</td>
<td>0.0048</td>
<td>0.2274</td>
<td>0.27</td>
<td>0.1266</td>
<td>0.099</td>
<td>0.0312</td>
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<tr>
<td>used DASC (mg/g)</td>
<td>36.654</td>
<td>0.021</td>
<td>12.624</td>
<td>3.2388</td>
<td>0.0042</td>
<td>1.7484</td>
<td>0.1434</td>
<td>37.176</td>
<td>0.1962</td>
<td>0.0618</td>
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<td></td>
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<td></td>
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<tr>
<td>Tot. conc (feed, mg/l)</td>
<td>0.16</td>
<td>0.05</td>
<td>87.99</td>
<td>0.9</td>
<td>0.05</td>
<td>14.96</td>
<td>0.1</td>
<td>15.32</td>
<td>IND</td>
<td>0.14</td>
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<tr>
<td>Tot. conc (eff, mg/l)</td>
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<td>0.05</td>
<td>15.69</td>
<td>0.35</td>
<td>0.05</td>
<td>4.53</td>
<td>0.35</td>
<td>0.86</td>
<td>IND</td>
<td>0.07</td>
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<td>Dissol. conc (feed, mg/l)</td>
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<td>0.004</td>
<td>85.99</td>
<td>0.028</td>
<td>0.001</td>
<td>21.99</td>
<td>0.001</td>
<td>13.45</td>
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<td>0.005</td>
<td>16.6</td>
<td>0.271</td>
<td>0.005</td>
<td>9.081</td>
<td>0.615</td>
<td>0.243</td>
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<td>0.152</td>
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<tr>
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<td>0.16</td>
<td>0.05</td>
<td>60.29</td>
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<td>0.05</td>
<td>11.57</td>
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<td>36.32</td>
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<td>0.05</td>
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<td>0.005</td>
<td>52.03</td>
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<td>0.001</td>
<td>16.88</td>
<td>0.252</td>
<td>159.5</td>
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<td>39.66</td>
<td>0.197</td>
<td>0.002</td>
<td>19.23</td>
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<tr>
<td>Tot. conc (feed, mg/l)</td>
<td>0.18</td>
<td>0.05</td>
<td>43.32</td>
<td>0.59</td>
<td>0.05</td>
<td>9.73</td>
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<td>268.2</td>
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<td>Tot. conc (eff, mg/l)</td>
<td>0.31</td>
<td>0.05</td>
<td>29.76</td>
<td>0.35</td>
<td>0.05</td>
<td>9.3</td>
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<td>136.1</td>
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List of Figures

Fig 1: Schematic diagram of the laboratory based DASC constructed wetland system

Fig 2: Treatment performances expressed as percentage removal efficiencies in the DASC based constructed wetland system for phosphorus (top) and BOD$_5$ and COD (bottom)
Fig 1 Schematic diagram of the laboratory scale DASC based constructed wetland system
Fig. 2 Treatment performances expressed as percentage removal efficiencies in the DASC based constructed wetland system for phosphorus (top) and BOD$_5$ and COD (bottom)