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Pilot field-scale demonstration of a novel alum sludge-based constructed wetland system for enhanced wastewater treatment

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Abstract

In this study, beneficial reuse of the alum-contained drinking water treatment sludge is extended into developing a novel constructed wetland system (CWs) using the alum sludge as main substrate. The study reports on the first pilot field-scale alum sludge-based CWs operated in the tidal flow mode with enhanced capacity for phosphorus and organic matter removal from animal farm wastewater. The concept of the development is presented and this is followed by the performance analysis of the first CWs of its kind. The CWs consists of four identical compartments in series operated using a tidal flow strategy with a hydraulic loading rate of 0.29 m$^3$/m$^2$.d. First year analysis of the system’s performance shows that it is a unique and promising low-cost wastewater treatment system. The mean monthly removal efficiencies obtained was determined to range from 57%-84%, 36%-84%, 11%-78%, 49%-93%, 75%-94%, 73%-97% and 46%-83% for BOD$_5$, COD, TN, NH$_4$-N, TP, P (inorganic phosphorus) and SS. The system showed a distinct phosphorus removal and also, the system was effective in reducing levels of organics and ammonium-nitrogen. More importantly, the system showcases a novel reuse alternative for the alum sludge as opposed to its landfilling, demonstrating a win-win technique with a great potential for larger-scale application.

Keywords Alum sludge; constructed wetlands; phosphorus; tidal flow; wastewater treatment.
1. Introduction  

Aluminium-based water treatment sludge (commonly referred to as alum sludge) is a by-product derived from the purification processes of drinking water treatment plants when aluminium (Al) salts are used as coagulant. It is the most widely generated drinking water treatment residual worldwide since Al-salts are the most widely used primary coagulant for water purification. Accordingly, alum sludge consists of various impurities in the raw water (e.g. colour, turbidity, hardness and varied concentrations of organics and microorganisms) and coagulant products and residues. Generally, in Ireland and most other places in the world, alum sludge is regarded as a “waste” and consequently buried as a waste material in landfills. However, alum sludge is predominantly composed of amorphous Al up to 29.7±13.3% dry weight [1] and these generally have larger surface area and greater reactivity toward anion adsorption than the corresponding crystalline mineral phases.

Due to the high reactivity of alum sludge and the strong chemical affinity of Al for phosphorus (P) in wastewater, alum sludge has huge potential for use as valuable material in wastewater treatment engineering. A number of researchers have demonstrated that alum sludge can be utilized in wastewater treatment to enhance adsorption and chemical precipitation processes that remove various pollutants (especially P) in wastewaters [2-5]. Furthermore, compared with other industrial by-products, alum sludge is unique in that it is a locally, easily and largely available by-product in most cities and metropolis worldwide. Therefore, the beneficial reuse of alum sludge, hitherto considered as a waste by-product for wastewater treatment represents an innovative approach using waste for wastewater treatment.

The authors’ research group has conducted extensive work to identify the characteristics and P adsorption capacity of alum sludge, which is generated from the largest drinking water treatment plant in Ireland. It has been demonstrated that the alum
sludge is a reliable and cost-effective material for enhancing P removal in wastewater treatment [6,7]. More significantly, a novel constructed wetland system (CWs) with enhanced capacity for P removal has been developed on a laboratory scale by employing alum sludge as main substrate [8, 9]. CWs have been recognized as one of the environmentally friendly technologies and they have been increasingly employed worldwide for the treatment of a diverse range of wastewaters. This paper reports on the first field study of a pilot-scale CWs utilizing alum sludge as the main substrate. The concept of developing the alum sludge-based CWs and a short recall of the development is firstly presented.

2. The concept

There is an urgent and universal need to develop innovative reuse options for inevitable industrial by-products in line with the environmental policy of “reduce, reuse and recycle” and sustainable development. Urbanization, industrial revolution, economic development and various daily life activities have polluted waters and left a legacy of sediments in the aquatic environment. Appropriate disposal of such resultant wastes, particularly in beneficial ways remains a great challenge to engineers and scientists. One of such inevitable industrial by-products is alum sludge which is being continuously generated on daily basis worldwide due to the priority need of water supply for human living. A critical review of alum sludge disposal showed that it is treated mainly as “a waste for landfill” with little known beneficial reuse value [1]. Therefore, development of alternative options for end-uses remains a great challenge in water and environmental research.

On the other hand, wastewater treatment including nutrients (N and P) removal is the target of municipal wastewater treatment plants. Except for the large scale activated sludge process, many treatment technologies have been extensively investigated. CWs
have demonstrated their advantages in terms of aesthetics, lower energy consumption and more economical construction and operation. The interest in the application of CWs has grown steadily over the years and it has now been applied to treat various types of wastewaters in virtually every continent [10]. In addition, CWs are also considered as the most attractive decentralized wastewater treatment system, which is technically appropriate and economically affordable [11]. Generally, CWs performance is good in terms of removal of organic matter and suspended solids, but as regards nutrients reduction, particularly P, the performance has been inconsistent and often low [11,12]. Consequently, one of the important research questions in CWs development lies in seeking novel materials to replace the normally used soil, sand and gravel in order to improve P removal, particularly through adsorption/immobilization [13-15]. Alum sludge is thus proposed as a novel low-cost substrate material for CWs due to its specific features of (1) high content of Al ion which plays a key role for P immobilization, thereby making the sludge a reliable and cost-effective adsorbent for P removal; (2) easy, large and local availability. Such reuse of alum sludge in a wastewater treatment stream is conceptually illustrated in Fig. 1. Clearly, it represents an attempt to use “waste” for wastewater treatment, making the alum sludge-based CWs a win-win technique and a contribution to sustainable development.

3. The first pilot-scale study

3.1 System set up, operation and monitoring

The pilot field-scale demonstration of the novel CWs was carried out on an animal research farm located at the Lyons Estate, Newcastle, Co. Dublin (53.3°N and 6.5°W), Ireland. The farm currently comprise of ca 17,000m² of farm and laboratory buildings with over 2,000 livestock units of sheep, pigs, cattle and horses. The farm is located close
to many development clusters and there is great attention being paid to the treatment of wastewater generated from the farm activities. The CWs was set up on the farm and it consists of four-stage down flow wetland cells. Fig. 2 shows the field set-up of the CWs and the development of the reeds from set-up up until the second and fourth month, respectively. Each stage of the CWs was constructed using similar 1100L plastic bins connected with submersible pump placed in each stage. A well was created in the middle of each stage and this housed the pump and also served for taking the samples.

[INSERT FIG. 2 HERE]

Each stage was configured using 10mm gravel at the bottom up to a depth of 10cm followed by 65cm of the alum sludge cakes as the main substrate layer and then 10cm of 20mm gravel to serve as the distribution layer. The alum sludge cakes used were collected fresh from the industrial filter press of the sludge dewatering unit of a drinking water treatment plant in Southwest Dublin, Ireland where aluminium sulphate is used as coagulant. Detailed characterization of the alum sludge in relation to its use as a substrate in a CWs has been published [7]. It should be re-emphasized that the CWs was operated in the tidal flow mode in which wastewater is periodically loaded and drained from each stage of the CWs in batches and the loading and draining operations are controlled by pumps. On loading (via pumping) of wastewater onto the top surface of the CWs, the wastewater quickly percolates/trickles down to fill up the pore spaces (occupy the pore volume essentially) and during the contact period, there is interaction with the alum sludge and biofilm developed on the surface of the alum sludge and these contribute to reducing the pollutant concentration in the wastewater. After the set period of contact, the wastewater occupying the pore volume is drained by the action of pumps which pumps out the wastewater. There was nothing unusual about this operation from our field
experience and the specific mode of operation adopted for the system served to allay any fears of problems with wastewater flow in such alum sludge based system. However, as it is normal in any wastewater treatment system, the gradual build up of solids on the surface of the system will eventually lead to clogging and this is inevitable. At this stage, the draining action via pumping may be affected, but there are quite a number of options for dealing with this e.g. scrapping/removing the top surface, bed resting, solubilisation etc. It should however be noted that clogging is not only peculiar to such alum sludge based constructed wetland system, rather it is typical of most wastewater treatment system.

Common reeds, *Phragmites australis*, were planted on top of each stage at the beginning of the experimental trials and good growth with lush vegetation was observed after two months. The system was operated as a subsurface flow system using a tidal flow operation strategy [16]. The “tides” of rhythmic filling and draining were generated by the pumps in each stage. Each pump was connected to a digital electronic timer which controls its operation and hence, the movement of wastewater from the influent tank and across the stages sequentially from the first to the last stage based on a programme schedule. Wastewater from the farm activities was firstly collected from the holding tank on the farm and pumped into a 10,000L capacity tank. Appropriate dilution was then carried out using tap water to achieve desired concentration which is conducive for the initial growth and establishment of the reeds. Accordingly, the influent wastewater into the system had a range of concentrations of BOD$_5$ (31-968 mg/l); COD (124-1634 mg/l); PO$_4$-P (2.8-60 mg-P/l); TN (16-273 mg-N/l) and SS (25-633 mg/l). Thereafter, the wastewater was gravity-fed into an underground tank (with a ball-float valve control) which serves as the influent tank from where the wastewater is pumped into the CWs.

There were 3 cycles per day and each cycle consists of 4 hours of wastewater contact in each stage and four hours of rest during which wastewater is drained out (to the next stage) and the stage is left to rest. A designed hydraulic loading rate of 0.29 m$^3$/m$^2$.d
(where \( m^2 \) represents the total surface area of the system) was applied. Samples of influent (from the underground influent tank) and effluent (from the four stages) were collected weekly and analysed for COD, BOD\(_5\) (Lovibond OxiDirect apparatus (Lennox, UK)), TP (Ascorbic method [17]), P (PO\(_4\)-P, inorganic phosphorus), TN (Persulfate method [17]), NH\(_4\)-N, NO\(_3\)-N, NO\(_2\)-N, SS, Turbidity (Hach turbidity meter 2100N IS) and pH (YSI multiprobe system, YSI incorporated, USA). Except where indicated, all the water quality parameters were analysed using a Hach DR/2400 spectrophotometer according to its standard operating procedures. To determine the pollutant removal efficiencies for the system, the amount (in terms of concentration) of pollutant removed is divided by the influent concentration and expressed as a percentage.

### 3.2 Results

Results and performance evaluation are based on data obtained between February and December 2009. Performance data (based on monthly averages) including the influent and effluent concentrations and the removal percentages are presented in Fig. 3(a) (for BOD\(_5\) and COD) and Fig. 3(b) (for TN and NH\(_4\)-N). Average pH (data not shown) showed a slight decrease between the influent (mean pH=7.6) and the final effluent (mean pH=7.3). The mean monthly influent concentrations varied appreciably and this was as a result of seasonal influence on farm activities which was reflected in the highly variable nature of the characteristics of the wastewater generated on the farm. Accordingly, the range of mean monthly influent concentrations (in mg/l) for BOD\(_5\), COD, TN and NH\(_4\)-N were 41.2-694.4, 407.0-1297.5, 43.0-221.9 and 37.9-176.2, respectively. The mean monthly removal efficiencies obtained was determined to range from 57%-84%, 36%-84%, 11%-78% and 49%-93% for BOD\(_5\), COD, TN and NH\(_4\)-N, respectively while the CWs achieved removal rates (in g/m\(^2\).d) ranging from 7.1-149.8(BOD\(_5\)), 49.8-253.8(COD), 7.1-47.0(NH\(_4\)-N), 0.9-38.3 (TN), 2.5-8.9 (TP) and 21.4-58.9 (SS).
The highest mean monthly removal efficiencies were obtained in August (for COD), September (for BOD$_5$ and NH$_4$-N) and October (for TN) while the least mean monthly removal efficiencies were obtained in February/March (for COD and TN), April (for BOD$_5$) and December for NH$_4$-N. Fig. 3(b) indicates that the system was very effective in reducing ammonium-nitrogen with a relatively stable final effluent concentration. By comparison, the system achieved a greater reduction for ammonium-nitrogen than for total nitrogen except for the month of December.

[INSERT FIG 3 HERE]

Fig. 4 shows the performance results for (a) TP and P and (b) SS and turbidity. Similarly, the system achieved excellent removal efficiencies for these pollutants and it further demonstrates the effectiveness and treatment potential of the system particularly for high strength wastewater treatment. At a range of mean monthly influent concentrations of 10.7-33.3, 8.6-32.8, 101.2-346.7 and 66.1-177.4 mg/l, a range of mean monthly removal efficiencies of 75%-94%, 73%-97%, 46%-83% and 36%-73% for TP, P, SS and turbidity, respectively, were obtained. A striking feature of this performance is that it showed a distinctive superior performance regarding P removal. Despite the high P loading onto the system, it sustained consistent high removal efficiency and this is attributed to the P removing ability of the alum sludge used as substrate in the system. High removal efficiency of SS was also observed particularly in the period between May and September. Generally, it can be observed from Figures 3 and 4 that the effluent concentrations mirror the influent concentrations which suggests that the system has a mass loading limit. It would therefore necessary to establish the loading limits for optimum efficiency.

[INSERT FIG. 4 HERE]
Fig. 5 shows the relative contribution of each stage (in percentage terms) to the overall removal achieved in the system for the different pollutants. Except for the removal of TN where the third stage had the highest contribution to the overall TN removal, the first stage accounted for the highest contribution to the overall removal for all the other pollutants monitored. Similarly, except for TN, the first two stages accounted for over 50% of the total removal achieved in all cases, and up to 75% for TP and P. However, the contribution of the third and fourth stages cannot be discounted as it varied depending on the pollutant. For instance, the third and fourth stages accounted for 59.8% of the overall TN removal compared to 24.6% and 24.4% for TP and P respectively. The pattern of BOD$_5$, COD and SS removal were similar in the first stages highlighting the influence of SS removal on the removal of organics. However, the patterns were dissimilar, especially in the case of BOD$_5$ for the third and fourth stages.

[INSERT FIG 5 HERE]

4. Discussion

This study is aimed at demonstrating the feasibility and efficiency of a novel tidal flow CWs integrating alum sludge as the main substrate for enhanced wastewater treatment. Results from the study indicate that the system could effectively reduce the output of BOD$_5$, COD, TN, NH$_4$-N, TP, P, SS and turbidity. In all cases, the treatment efficiency obtained in the novel system was comparable to, or higher than, the performances obtained in conventional and other similar wetland systems. Sun et al. [18] noted that although the removal of organic substances (typically 80–99%) is now satisfactory in most CWs, the typical percentage of ammonia-nitrogen removal during long-term operation is 30%–50%. In a survey of 51 vertical sub-surface flow CWs, Vymazal [19] reported a mean removal efficiency of 44.6% for TN removal. Furthermore, Sun et al. [20] reported mean removal efficiencies of 97.6% and 71.3% for BOD$_5$ and COD respectively, for a
combined tidal-flow down flow reed bed treatment system with multiple sub-beds. However, the sub-beds have surface areas ranging from 2.56 m$^2$ to 3.25 m$^2$ each and this would result in lower overall loading (as compared to the loading used in this study). In other similar studies, Cerezo et al. [21] reported removal efficiencies of 90% and 70% for BOD$_5$ and COD, respectively, for a field based CWs made of eight tanks in three series and with a surface area of 1m$^2$. While the system in this study had a relatively smaller footprint and higher loading, it can be seen that the performance obtained are comparable and promising.

The system also showed good nitrification performance. This is believed to be due to the tidal flow strategy which enhances the oxygen mass transfer and thus leads to a greater capacity for microbial degradation. The pH values of the wastewater did not change appreciably across the system and mean values of 7.6, 7.5, 7.6, 7.5 and 7.3 were obtained for the influent and stages 1, 2, 3 and 4, respectively. Interestingly, the individual contribution of the different stages to the overall removal of ammonium-nitrogen was 35.8%, 27.8%, 25.7% and 10.7%, respectively, indicating that the first three stages accounted for ~90% of the total removal. Furthermore, whereas ammonium-nitrogen was reduced across the four stages, both nitrite- and nitrate-nitrogen increased across the stages (data not shown). The increases of nitrite- and nitrate-nitrogen are the results of nitrification as the ammonium-nitrogen is oxidised into nitrite- and nitrate nitrogen.

In relative terms, the removal efficiencies obtained in this study can be considered as excellent and showing good promise for the novel CWs based on two reasons (i) the performances were obtained during the first year of operation of the system and it also includes a 3-months start up and acclimatization stage. Yet, the results obtained are comparable and in some cases, showed better treatment results when compared to similar systems; (ii) based on the influent BOD$_5$ concentrations and daily flow rate used, the operation of the system translates into a design sizing of about 0.5 square metres per
population equivalent (0.5m²/pe) which is a fraction of the general recommended design
guides of 2m²/pe to 20m²/pe found in literature for sizing vertical flow CWs [10, 22]. It
should however be noted that the recommended design guides are mainly for secondary
systems treating domestic wastewater and there is no consensus yet available in literature
for systems treating high strength/industrial wastewater.

The system proved very effective P removal. In CWs, it is often a challenge to achieve
concurrent high removal efficiencies for P and organic matter (BOD₅, COD). Often times,
efficiency of P removal is usually low compared to other parameters such as BOD₅ [11].
In a pilot-scale study involving hybrid and integrated constructed wetland systems, Lee et
al. [23] reported BOD₅ reduction ranging from 38 to 65% but P concentration was merely
reduced, with reduction ranging from 8.6 to 13.1%. This is also similar with results
obtained by Stefanakis et al. [24] who reported removal efficiencies of 71.1%, 66.9%,
36.9% and 37.9% for BOD₅, COD, TP and P respectively. The result indicates that while
the removal of organics was satisfactory in their study, concurrent satisfactory removal of
TP and P was not achieved. It can thus be seen that significant and high P removal can be
obtained concurrently with high removal of organics in CWs by using alum sludge as the
substrate in a tidal flow CWs.

Although it is argued that P removal by adsorption onto the substrate media has a finite
capacity and does not contribute to the long-term sustainable P removal, it is interesting to
note that the use of alum sludge as a substrate in such CWs can significantly extend the
service lifetime of the CWs. The use of a material with high adsorption capacity, as alum
sludge, can significantly extend the life time of the CWs, but according to actual
knowledge sooner or later the performance of the system is expected to decrease as a
result of system saturation, and so a longer time study is recommended. In a previous
laboratory-scale study by the authors, it was determined that the alum sludge-based CWs
can have a service lifetime of 4-17 years (based on P removal) and this is significantly
longer than that for typical CWs [8]. Therefore the alum sludge-based CWs is a unique and promising low-cost wastewater treatment system particularly for isolated or scattered settlements, agricultural and industrial effluents, private dwellings, hotels, parks, rural areas, etc. Simultaneously, the system also showcases a novel reuse alternative for the alum sludge as opposed to its landfilling, demonstrating a win-win technique. Perhaps as water companies now strive to include sustainability principles in their business practise, this unique feature of recycling alum sludge will be very strategic. However, prior to this, longer term operation of the field system is desirable and this is currently been undertaken. Such longer term operation will give more opportunity to carry out further research on the mechanism and life span of this new kind of CWs, especially for TP removal and this would complement similar laboratory scale investigation that has been carried out by the authors previously [6].

5. Conclusions
The effectiveness of a novel alum sludge-based CWs was demonstrated through one year performance analysis of pilot field-scale system. Results obtained showed that the system holds great promise as a low-cost wastewater treatment system for enhancing pollutants removal efficiencies, especially P. At a range of mean monthly influent concentrations (in mg/l) of 41.2-694.4, 407.0-1297.5, 43.0-221.9, 10.7-33.3 and 37.9-176.2 for BOD₅, COD, TN, TP and NH₄-N, respectively, mean monthly removal efficiencies of 57%-84%, 36%-84%, 11%-78%, 75%-94% and 49%-93% were obtained. While excellent removal efficiencies were achieved in the system, at the same time, the system offered a novel reuse alternative for the alum sludge as opposed to landfill. It is expected that with a continuing period of operation, vegetation and microbial population will become well established leading to a sustained and even improved treatment performance.
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References


**Figure captions**

**Fig. 1** Conceptual illustration of beneficial integration of alum sludge into a wastewater treatment

**Fig. 2** Field set-up of the constructed wetland system and reeds development

**Fig. 3** Mean monthly influent and effluent concentration and removal efficiencies for (a) BOD$_5$ and COD and (b) TN and NH$_3$ (The bars represent the removal efficiencies while influent and effluent concentrations are shown as line plots)

**Fig. 4** Mean monthly influent and effluent concentration and removal efficiencies for (a) TP and P and (b) SS and Turbidity (The bars represent the removal efficiencies while influent and effluent concentrations are shown as line plots)

**Fig. 5** The relative contribution of each stage (in percentage terms) to the overall removal achieved in the system for the different pollutants.
Water treatment plant

Water supply

Wastewater

Alum sludge-based constructed wetland system for wastewater treatment

River

Alum sludge (byproduct)