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From “fills” to filter: Insights into the reuse of dewatered alum sludge as a filter media in a constructed wetland

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Abstract
Dewatered alum sludge, a by-product of drinking water treatment plants, hitherto consigned to landfills was used to develop a novel bio-filter in form of a constructed wetland. Performance results have demonstrated the benefits of the alum sludge cakes in a lab-scale system in enhancing phosphorus (P) removal from an animal farm wastewater. Although P and organic matter were concurrently removed in the system, there was a probable “one off” release of organics from the system, and this coincided with an increase in inlet P concentration from 39.2 mg-P/l to 163.0 mg-P/l. A conceptual model was then proposed to explain and discuss this.

Keywords. Adsorption, dewatered alum sludge, phosphorus, vertical flow constructed wetland, waterworks sludge, wastewater treatment, organic matter.

Introduction

Drinking water treatment generally includes a coagulation/flocculation stage during which particulate, colloidal and soluble contaminants are entrapped or adsorbed on coagulant flocs and subsequently removed from the liquid phase during sedimentation. In most water treatment plants around the world, aluminium sulphate is the most widely used coagulant and accordingly, the residual sludge obtained is termed “alum sludge”. Alum sludge has a high moisture content of about 98% or more (Cheremisinoff, 1994) and dewatering is often employed to reduce the bulk water content. This results in a dewatered alum sludge cake with a variable solid content. The dewatered alum sludge cake has high content of amorphous aluminum and its appropriate disposal is now a significant environmental issue. Although disposal into licensed landfills or dedicated mono-fills has been the most common method for disposing the dewatered alum sludge, such approach fails to recognize the intrinsic potential benefits of reusing the aluminium content embedded in the sludge. In addition, policy restrictions, increasing landfill tax and costs and the perceived potential harmful effect of the aluminium content in the sludge have all combined to necessitate increased research into suitable alternative reuses or disposal routes for the alum sludge.

In the past, alum sludge cakes have been utilized as a component in the manufacture of several materials such as concrete, cement mortars, clay materials and fired ceramic products (e.g. bricks, pipes and tiles); as geotechnical works material; as a potential material for use in silviculture and agriculture and for phosphorus (P) removal from aqueous solutions (Babatunde and Zhao, 2007). However, giving the inevitability of alum sludge generation at drinking water treatment plants and the escalating costs and concerns associated with its landfill disposal, continually seeking alternative disposal routes becomes imperative. Even though among other potential uses, dewatered alum sludge cakes have been shown to immobilize P in aqueous solutions, there is no reported/published study yet, that extends such ability of the sludge as a main/raw material for wastewater treatment. It is therefore proposed to develop a novel bio-filter in

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form of a constructed wetland system using the dewatered alum sludge cakes as the main substrate.

Constructed wetland systems are low-cost wastewater treatment systems that are now recognized and used all over the world (IWA, 2000). However, since the traditional wetland systems employ gravel and/or local soils as the substrate, P removal is often poor in these systems due to the limited P removal capacity of the gravel and/or the local soil used. As a remedy, it is often suggested to use substrate materials that are rich in aluminium and iron, considering the fact that these metal ions have a strong affinity for P (orthophosphate) in aqueous solution (Lena, 2006). In tandem with this line of thought, it is hoped that the use of dewatered alum cakes as a substrate media would enhance P removal due to its high content of amorphous aluminium. To contribute to the successful development of the system, this paper explores in part, the lab-scale performance of such system, focusing on the concurrent removal of phosphorus and organic matter with an attempt to gain insight into the processes occurring.

Materials and methods

Dewatered alum sludge cakes designated for landfills were collected from the biggest water treatment plant in Ireland. The pH of a 0.01M $CaCl_2$ solution of the sludge cake (particle size < 2mm) was determined according to the procedures outlined by McLean (1982) using a 1:10 solid to liquid ratio. Water content of the sludge was determined by oven drying at 103^{o}C. Loss of ignition (LOI) was determined in accordance with the British Standards (British Standard Institution, 1990). Bulk density was determined from the volume of water displaced by a known mass of the sludge sample. The elemental composition of the sludge was also carried out and this has been reported elsewhere (Yang et al., 2006). To examine the proposed use of the sludge in a bio-filter/constructed wetland, two lab scale vertical flow unvegetated constructed wetland systems (VFCWs) were constructed using pyrex tubes (1.0m long with an internal diameter of 9.5cm). Fig 1 shows schematic description of the systems. The lab scale constructed wetlands were packed with air-dried sludge and fed with diluted agricultural wastewater, collected from the secondary holding tank of an agricultural research farm, with a herd size of about 2000. One of the VFCWs (vf1) was operated intermittently in downflow mode while the second (vf2) was operated continuously in an upward-flow mode. The intermittent operation in vf1 involved periodically filling and draining the wetland with wastewater and a pre-set digital electronic timer regulated this. In vf2, the flow was continuous upward-flow and this was realised by pumping wastewater to the base of the vf2 through a vertical pipe that penetrated the alum sludge medium forcing the wastewater up through the medium. The two lab scale wetlands were fed with the wastewater from the same feed tank. Table 1 shows the operating conditions at the different operational phases of the experiment. Levels of soluble reactive P (SRP) and COD were monitored concurrently over a 24 weeks period.

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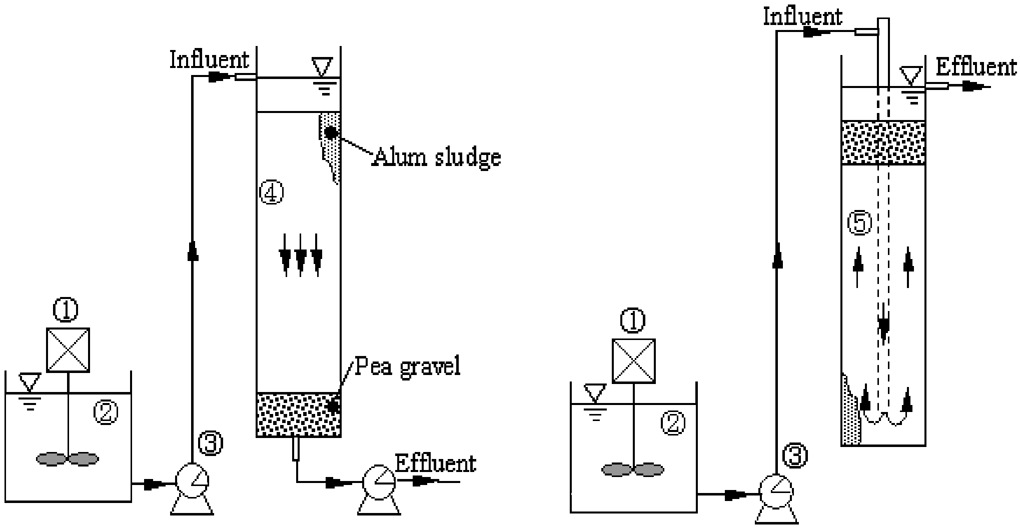

Fig.1 Schematic diagram of the lab-scale wetland systems: (1) Stirrer (2) Diluted agricultural wastewater (3) Peristaltic pump (4) & (5) Arrows indicating direction of flow.

Table 1 Wetlands operating conditions at different experimental phases

Parameter		Operating period					
		I	II	III	IV	V	VI
Contact time (hours)		3	3	4	4	4	4
Duration (weeks)		3	3	3	3	7	5
HLR (m^3/m^2.d)	Vf1	1.86	1.86	1.40	1.40	1.40	1.40
	Vf2	1.71	1.71	1.29	1.29	1.29	1.29
P-loading (g-P/m^2.d)	Vf1	15.30	35.30	54.88	54.88	228.2	456.4
	Vf2	14.0	32.50	50.57	50.57	210.3	420.5

Results

Selected physical and chemical characteristics of the sludge are presented in Table 2. The sludge has a circumneutral pH of 6.02 with a 74-75% water content. The pH is within the range of 5.10 to 8.00 as reported by Basta et al., (2000). The circumneutral pH of the sludge may however reflect the strong hydrolysis potential of the sludge. The sludge has an apparent high percentage of organic matter as indicated by the 59.8% LOI. Bulk density was determined to be 1.3g/cc. Aluminium (expressed as alum oxide) accounts for up to 46% of the dewatered sludge cake by mass while iron (expressed as iron oxide) accounts for about 1.2% (Yang et al., 2006). The high level of aluminium is quite expected as the aluminium sulphate used during the flocculation/coagulation stage may have formed a larger part of the flocculated mass removed after clarification. Typically, mean aluminium concentration (in % dry weight) of aluminium based water treatment works sludge is 29.7$\pm$13.3 (Babatunde and Zhao, 2007).

Table 2 Selected characteristics of the dewatered alum sludge

pH	Water content (%)	LOI (%)	Bulk density (g/cc)	Al (mg/g)*	Fe (mg/g)*
6.02	74-75	59.8	1.3	463	12.3

* From Yang et al., (2006)

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Fig. 2 shows the time course variation of treatment performance in the wetland systems. Even though P loading onto the wetlands was periodically increased, efficient P removal was obtained in both wetlands. There was no clear difference in P removal efficiencies between the two wetlands up to the 12th week and there was only a slight difference in the removal efficiencies as the end of the 24th week. The mean P removal efficiencies were 81% and 71% in vf1 and vf2 respectively as at the end of the 24th week. It should be noted that P loading at the end of the sixth operational period (24 weeks in total), was 420.5 g-P/m^2.d corresponding to an average inlet P concentration of 320mg-P/l.

Removal of COD increased gradually over time, with up to 60% removal efficiency obtained in both cases. However, a negative removal efficiency (i.e effluent COD value was greater than influent value) started to occur in both wetland columns from the 10th week in vf2 and from the 13th week in vf1. This was coincidental with an increase in inlet phosphorus concentration (especially during the 13th week) from 39.2mg-P/l to 163.0mg-P/l. Negative removal efficiency up to –336.3% was recorded in the systems. Even though inlet P concentration was further increased to about 326mg-P/l, no further negative removal was observed. This observed trend spurs up many interesting points and in order to probe this finding, a conceptual model was proposed.

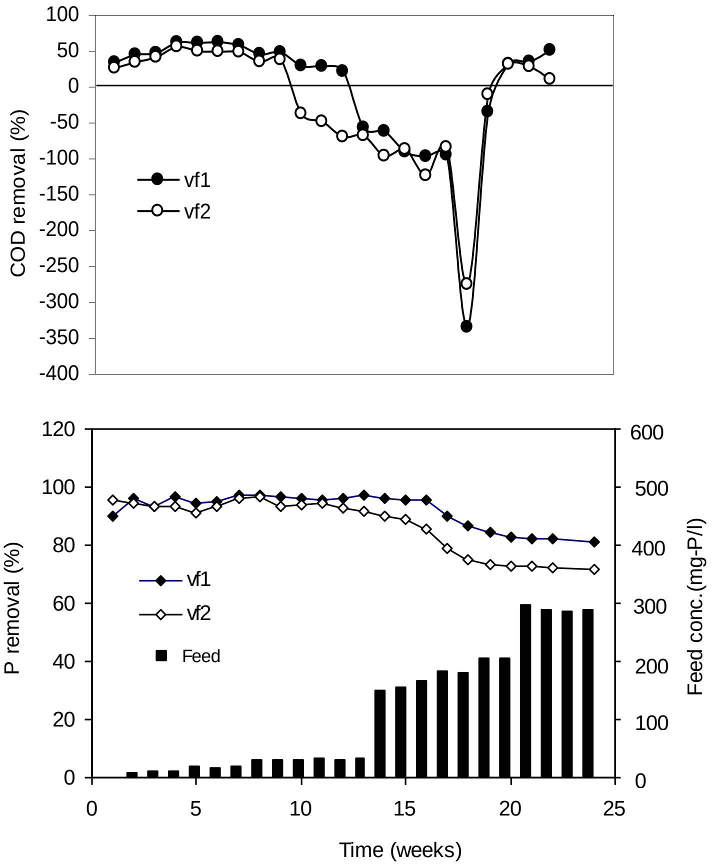

Fig. 2 Time course variation of treatment performance in the two lab-scale wetlands

Discussion

Performance data (Fig. 2) shows that efficient P removal can be obtained in both wetlands even at increased loadings. The efficient P removal obtained in the wetlands could be attributed to the amorphous aluminium content of the alum sludge cakes used as

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substrate in the wetlands. Numerous studies have demonstrated that aluminium ions have a strong affinity for P adsorption (orthophosphate) in aqueous solution (Lena, 2006; Yang et al., 2006; Razali et al., 2007). Noting that the sludge cakes were hitherto consigned to landfills, their use as substrates in a wetland could therefore be seen to have the potential to enhance P removal. Lena (2006) discussed several materials that have been tested in laboratory studies as potential substrates to enhance P removal. In comparison to most of the materials tested and the testing conditions (especially the P loading) it can be inferred that the alum sludge have a significant removal capacity for orthophosphate in wastewaters.

Regarding COD removal the initial removal of up to 60% was obtained in both cases. However, as shown in Fig 2, negative removal efficiency was observed in both wetlands especially from the 13th week when influent P was increased. It is an unusual trend and efforts have been made to try to understand what happens. Monitoring of soluble COD **(**sCOD**)** was conducted and the data shows a very limited removal (data not included), suggesting that the main mechanism for organic removal in the wetlands under investigation may have been through the filtration of suspended organics (expressed as particulate COD, pCOD) onto the surface of the alum sludge. Keeping in mind of this, the ensuing negative removal of COD with sustainable removal of P may reflect the competitive adsorption between initially adsorbed organics and continuous input of P. Kaiser et al., (1999) reported that increasing solution concentration of SO_3^{2-} and H_2PO^{4-} resulted in increasing organic carbon release from a soil material. In a similar study, Reemtsma et al., (1999) subjected surface soils from two former wastewater infiltration sites to release test using three salt solutions ($CaCl_2$, $NaNO_3$, NaH_2PO_4) at a concentration of 50Mm. It was found that the dissolved salts have a strong effect on the amount of organic matter released from the two soils with NaH_2PO_4 inducing the highest release of organic matter. Therefore, a model was proposed and shown in Fig. 3 to explain the possible mechanism of multiple sorptions of P and organic matter onto the surface of the alum sludge and the possible release of initially adsorbed organics at high P concentration. During wastewater contact with the active alum sludge surface, P (orthophosphate) is rapidly removed from the wastewater and adsorbed onto the alum sludge surface through the ligand exchange mechanism as identified and explained in detail in Yang et al., (2006). The filtered pCOD then firstly undergoes hydrolysis by bacterial action through which the particulate organic matter is converted from solid to dissolved phase (solubilized). Gujer et al., (1999) noted that particulate organic biodegradable substrates and high molecular weight dissolved and colloidal constituents must undergo cell external hydrolysis before they are available for biodegradation. Thereafter, the bacteria utilize the dissolved organic matter (DOM) and the dissolved oxygen (DO) concentration in the wetland determines the extent, and rate of the organic matter oxidation by bacterial action and also the final products formed.

Redox readings and DO measurements obtained during the 21st week from the two wetlands indicate that anaerobic condition prevails in the two wetlands with vf2 in a more anaerobic condition. There was also a mild olfactory detection of hydrogen sulphide gas and this also lends credence to the anaerobic state of the two wetlands. As the degradation action of anaerobic bacteria is much slower than aerobic bacteria, there is a possibility of incomplete utilization of the DOM. Therefore, concurrent with an increase in the P concentration in the wastewater and as more P is preferentially adsorbed onto the alum sludge, some of the initially adsorbed organic matter (including those that are yet to be utilized by bacterial action) may have been displaced and released into the

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effluent. This may have contributed to the negative COD removal efficiency. Overall, it is reasonable to infer that although competition from other negative ions and biofilm development did not have any negative effect on P removal, a high P concentration may have induced a "one off" release of initially adsorbed organics and this necessitates further research in order to ensure the sustainability of the proposed bio-filter.

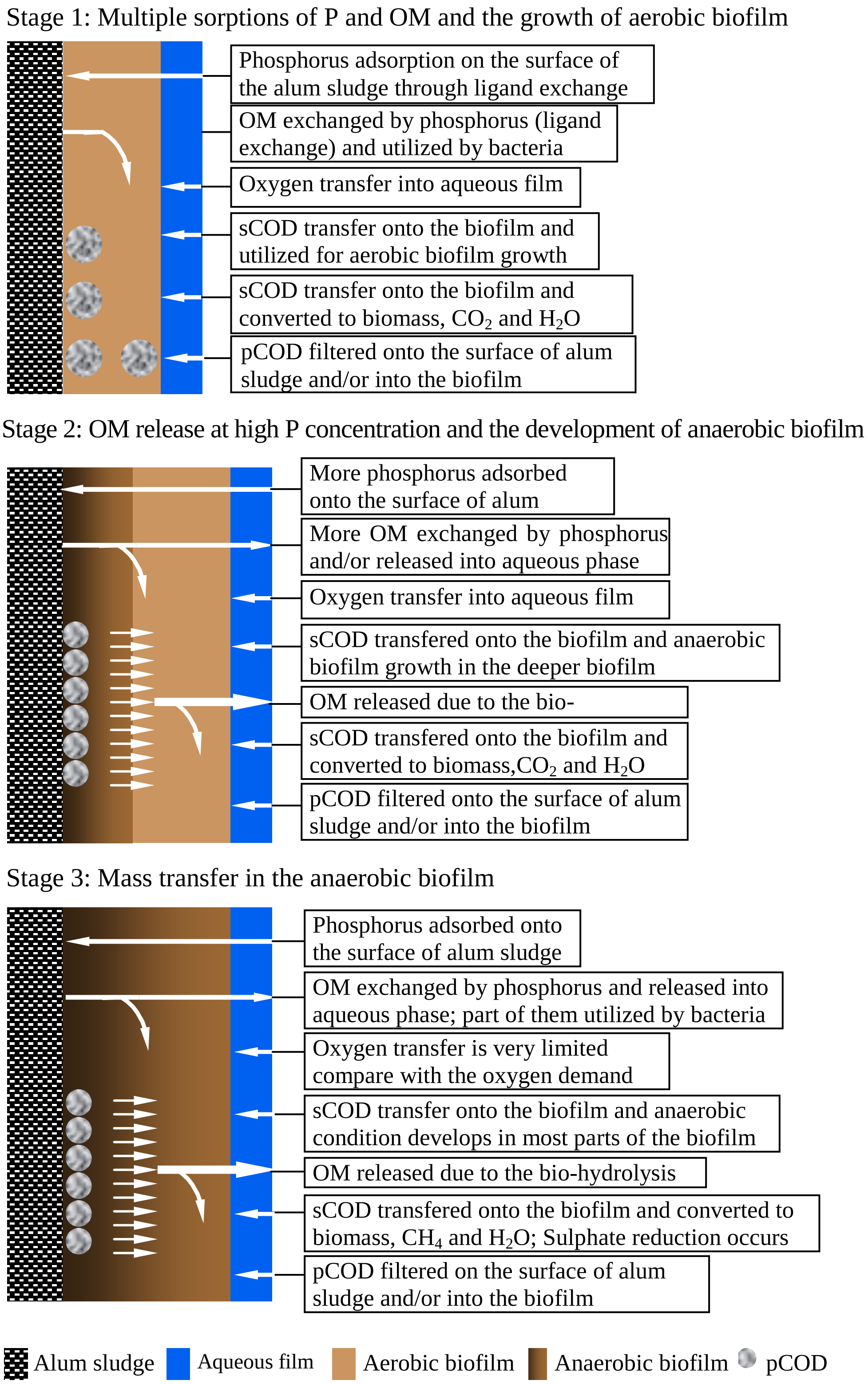

Fig. 3 Proposed conceptual model of multiple sorptions of OM and P onto alum sludge

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Conclusion

An alum sludge based bio-filter in form of a constructed wetland was developed with two main objectives: (1) To transfer the sludge from a “waste to landfill” material into a bio-filter material and (2) To utilize the sludge in a constructed wetland for the purpose of enhancing P removal. Results have shown that efficient P removal can be achieved and sustained in such an alum sludge based constructed wetland, irrespective of the design configurations. There was however a “one off” release of organics which coincided with an increase in inlet P concentration to about 320mg-P/l.

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