National Development Plan 2007 - 2013

Environmental Protection Agency
An Ghníomhaireacht um Chaomhnú Comhshaoil

STRIVE
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Environmental Technology: Development of an Alum Sludge-Based Constructed Wetland System for Improving Organic Matter and Nutrients Removal in High-Strength Wastewater

STRIVE

Environmental Protection
Agency Programme

2007-2013

SSTI
Strategy for Science, Technology and Innovation

Comhshaol, Oidhreacht agus Rialtas Áitiúil
Environment, Heritage and Local Government

Environmental Protection Agency

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EPA STRIVE Programme 2007–2013

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STRIVE Report

Prepared for the Environmental Protection Agency

by

University College Dublin

Author:
Yaqian Zhao

ENVIRONMENTAL PROTECTION AGENCY
An Ghníomhaireacht um Chaomhnú Comhshaoil
PO Box 3000, Johnstown Castle, Co.Wexford, Ireland

Telephone: +353 53 916 0600 Fax: +353 53 916 0699
Email: info@epa.ie Website: www.epa.ie

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The EPA STRIVE Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.

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Details of Project Partners

Yaqian Zhao
Centre for Water Resources Research
School of Architecture, Landscape and Civil Engineering
University College Dublin
Philips Building, Belfield
Dublin 4
Ireland
Tel.: +353 1 7163215
Email: yaqian.zhao@ucd.ie

Akintunde Babatunde
Centre for Water Resources Research
School of Architecture, Landscape and Civil Engineering
University College Dublin
Philips Building, Belfield
Dublin 4
Ireland
Tel.: +353 1 7163209
Email: akintunde.babatunde@ucd.ie

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Executive Summary

The generation of alum sludge in Ireland is inevitable. Its disposal, however – amidst decreasing available landfill space coupled with escalating costs and public concerns – remains an environmental quagmire. Seeking cost-effective and environmentally sustainable disposal alternatives is, as a result, a significant environmental issue. Alum sludge is a by-product of drinking-water-treatment processes in Ireland. It is obtained when aluminium (Al) salts are used as a chemical coagulant. Once alum sludge is dewatered, however, aluminium hydroxides become the dominant constituent, making it possible to reuse the sludge as a valuable raw material in wastewater treatment. This is because the ions enhance adsorption and chemical precipitation processes that remove various pollutants, especially phosphorus, from wastewater. Moreover, there is huge potential in Ireland for the application of small wastewater treatment systems, especially constructed wetlands (CWs), because of the unique geographic distribution of Irish residents. Constructed wetlands (commonly known in Europe as 'treatment reed beds') are regarded as a low-cost promising technology for wastewater treatment and have been increasingly employed for wastewater treatment since the early 1990s.

This project conceptualised and developed a novel CW system integrating alum sludge as its main substrate. The system has a multiprong feature of creating an alternative reuse option for alum sludge (as opposed to landfilling it) and, at the same time, enhancing wastewater treatment with high rates of phosphorus (P) removal. The system can be applied successfully to treat wastewater in various situations in Ireland, including individual houses, farms and scattered settlements. Furthermore, the concept of reusing alum sludge for P immobilisation introduced in this study can be integrated into any form of CW in Ireland to enhance P removal. The results obtained both in the laboratory and on the field have shown that the alum sludge-based CW system is a promising new technology that is green, low cost and environmentally friendly. Moreover, the results also confirm that the system can concurrently enhance the removal of P and organic matter from wastewater with very high removal efficiencies. Data obtained from the laboratory-scale trials indicate that average removal efficiencies for 5-day biochemical oxygen demand (BOD_5) (90.6%), chemical oxygen demand (COD) (71.8%), reactive phosphorus (RP) (93.3%) and soluble reactive phosphorus (SRP) (97.6%) were achieved. A similar treatment performance was obtained from the field trials of the system after one year of operation. At a daily wastewater load of 990 L, a range of mean monthly removal efficiencies of 57% to 84%, 36% to 84% and 73% to 97% were achieved for BOD_5, COD and SRP respectively.

Overall, the results have shown that alum sludge is (i) a good low-cost adsorbent for P immobilisation; (ii) can be a good support for biofilm development, providing a good environment for organics degradation; and (iii) is a suitable growth medium for the common reed. Environmental and health concerns regarding the reuse of the alum sludge were also addressed. Results reveal that the Al in the sludge could be released into the treated effluent, but, in most cases, the Al level in the leachates was below the limits of 0.2 mg/L as per European and international guidelines, and the release does not appear to pose an imminent ecological or environmental risk. It was also shown that P can be recovered from the saturated alum sludge and a recovery methodology was established.

1 Introduction

1.1 Alum Sludge and its Concerns

Sludge from water-treatment is one important by-product of the water industry. Aluminium-coagulated water treatment sludge ('alum sludge') is an inescapable by-product obtained as a result of water-treatment processes, such as coagulation, flocculation, clarification and filtration using aluminium sulphate as the primary coagulant (see Fig.1.1). The sludge that arises contains removed colour, turbidity and humic substances from source water, plus aluminium sulphate residues and polymers from the sludge-conditioning stage. The sludge is then dewatered. Because aluminium sulphate is the most widely used coagulant worldwide for source-water purification, alum sludge is the most widely generated water-treatment by-product.

In Ireland alone, an estimated 15,000 to 18,000 tonnes of dried solids of alum sludge are produced annually, and this is expected to double by the end of the next decade. In most countries, including Ireland, alum sludge is currently treated as a waste and it has become mandatory for water companies to dispose of the dewatered sludge appropriately. Most sludge is disposed of into landfills. The increasing amounts of alum sludge produced daily are triggering considerable environmental and economic concerns in addition to disposal issues. However, the options for alum sludge recovery/recycling are generally not well developed as economically viable options for application in Ireland. Thus, the search for cost-effective and eco-friendly disposal option(s) becomes an urgent priority.

1.2 Constructed Wetlands and the Need for their Further Development

Approximately 82% of Irish urban wastewaters receive secondary treatment. Among these treatment facilities, there are 144 constructed wetlands (CWs) under operation (Babatunde et al., 2008a). This means that CWs make a significant and measurable contribution to Irish environmental pollution control. The CW system is recognised widely as a 'green' wastewater treatment technique because of its low-energy requirement and aesthetic appearance. Constructed wetlands encompass a variety of treatment modules – including biological, chemical and physical processes, which

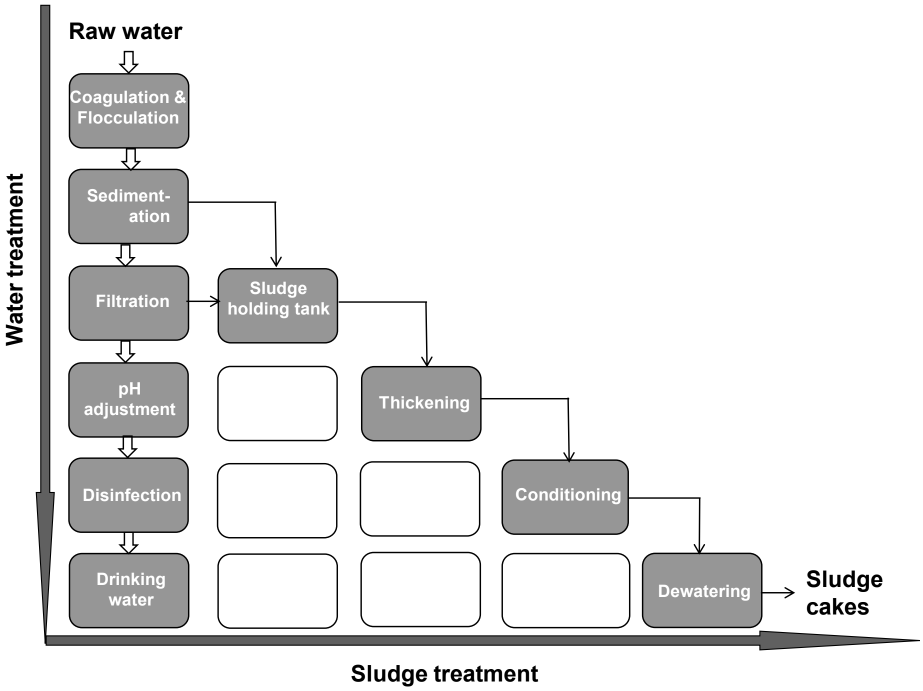


Figure 1.1. Typical water-treatment and sludge-treatment processes in a water-treatment plant.

are all akin to processes occurring in natural treatment wetlands. Theoretically, the treatment of wastewater within a CW occurs as it passes through the wetland medium and the plant rhizosphere. The decomposition of organic material is facilitated by the presence of aerobic and anaerobic microorganisms. Nitrogen removal is achieved mainly by microbial nitrification and denitrification while P is co-precipitated with iron, aluminium and calcium compounds located in the root bed medium. The plants (e.g. common reeds) also contribute to oxygen transfer within the system. Constructed wetlands have been increasingly used globally to treat various wastewaters (Kadlec and Wallace, 2009; Vymazal and Kröpfelová, 2009). Their performance is generally good for removing organics (termed COD & BOD_5) and suspended solids (SS), but inconsistent and often low in reducing nutrients (IWA, 2000). There is even more concern when CWs are employed to treat medium- to high-strength wastewater with high nutrient (especially phosphorus [P]) concentrations. This requires alternative substrates (rather than soil, sand, gravel and crushed stone, etc., which are more typical) with a high P sorption capacity in order to reduce P concentration to acceptable levels. In addition, when treating high-strength wastewater, it is necessary to enhance the oxygen-transfer efficiency in the CW to improve the organics removal.

1.3 Reuse of Alum Sludge in a Constructed Wetland: A Solution with a Two-Prong Feature

Globally, there is a need to reuse industrial by-products in line with moves towards sustainable development and environmental policies of ‘reduce, reuse and recycle’. This research examined the possible reuse of the Irish dewatered alum sludge as a low-cost alternative substrate in CW, particularly for P removal. This was based on the premise that alum sludge is predominantly composed of amorphous Al ions (29.7±13.3% dry weight [Zhao et al., 2006; Babatunde and Zhao, 2007]) and its ions enhance adsorption and chemical precipitation processes that remove various pollutants, especially P (Arias et al., 2001; Arias and Brix, 2005; Drizo et al., 1999; 2002; 2006; Johansson Westholm, 2006; Ago et al., 2008; Philip et al., 2009). In addition, alum sludge is relatively clean with respect to heavy metals and organics (Geertsema et al., 1994) and it is easily, locally and widely available. Therefore, the use of alum sludge as a substrate in CWs can bring about significant cost savings – in terms of its disposal and also in terms of wastewater treatment. This type of reuse of alum sludge in a main wastewater treatment stream is illustrated in Fig. 1.2. Obviously, it is an attempt to use ‘waste’ for wastewater treatment, demonstrating a win-win technique.

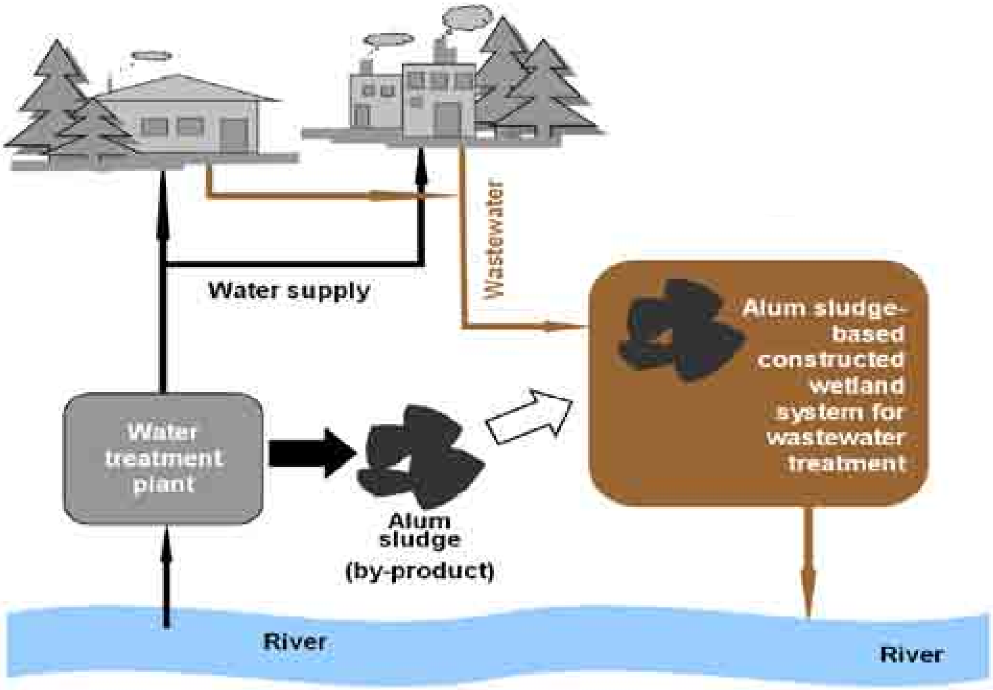


Figure 1.2. Conceptual illustration of beneficial reuse of alum sludge in wastewater treatment.

1.4 Objectives

This research project aims to develop a novel alum sludge-based CW system for high-strength wastewater treatment. Its novelty lies in the use of dewatered alum sludge as a substrate in the CW system. Specific objectives of the project are to:

- Investigate the feasibility of using dewatered alum sludge as the main medium of CW to treat high-strength nutrient-rich wastewater, with the main focus on the removal of organics (BOD_5, COD), P and ammonium nitrogen (NH_4-N) from the wastewater;
- Conduct a series of experiments in laboratory-scale and then pilot-scale wetlands planted with the common reed, *Phragmites australis*, and filled with different proportions of the dewatered alum sludge. Monitor the removal efficiency of BOD_5, COD, P and NH_4-N;
- Test the so-called ‘tidal flow’ operation strategy and determine the appropriate configuration of a multi-stage CW system based on the pollutants-removal performance;
- Study the mechanisms and kinetics for the removal of BOD_5, COD, PO_4-P and NH_4-N in the proposed novel wetland system;
- Investigate the possible release of substances, such as Al ions and more importantly polymer residuals, from the alum sludge cakes when the sludge is used as the main substrate in wetlands. This will provide vital information on the potential environmental impact of the reuse of alum sludge;
- Provide preliminary design and operation information for possible large-scale and industrial practices of this type of wetland treatment system.

The research work was carried out in several phases, including initial static bottle adsorption tests; an alum sludge column configuration trial; single alum sludge bed and wetland tests; a multistage alum sludge wetland system trial; and a field pilot-scale trial. Initially, the focus was on assessing the feasibility of using the dewatered alum sludge as an adsorbent, and its capacity for P adsorption. Thereafter, the optimal configuration of the proposed alum sludge-based CW system to give the best operational, hydraulic and treatment performance was determined using four different model reed beds systems. Here, the focus was on establishing the magnitude of the overall pollutant removal in the different model systems. Once the optimal configuration of the reed bed system was established, a tidal flow model CW with dewatered alum sludge as a main substrate was operated on a short- and long-term basis. At the same time, a methodology for monitoring polymer residual and possible release from the reuse of the dewatered alum sludge cakes in CW was also developed. Thereafter, a multistage CW system employing 100% alum sludge as the substrate was designed and continuously operated to treat agricultural wastewater under various organic loadings and operating conditions. The study of this phase also focused on addressing the lifetime of the alum sludge-based CW system and possible P recovery when the sludge becomes fully saturated. Finally, field trials of the pilot-scale systems derived from the laboratory-scale development were conducted on an animal farm as a demonstration study for further development. The potential for wetland clogging and the possible release of substances/elements (especially Al) from the alum sludge into the treated wastewater upon passage through the system was also examined.

2 Profile of Alum Sludge and Current Status of Constructed Treatment Wetland: Literature Review

2.1 Constructive Approaches toward Alum Sludge Management

The available literature estimates that, globally, 10,000 tonnes of water-treatment sludge are produced daily (Dharmappa et al., 1997). The costs of handling the enormous quantities of sludge account for a significant part of the overall operating costs of water-treatment works, and they are likely to increase because of increasingly stringent regulations. As noted by Viraraghavan and Ionescu (2002), the limited land available for alum sludge disposal and the possible environmental liabilities that may arise if it is disposed of in sanitary landfill sites make this a considerable worry for water-purification authorities.

On the other hand, it has been advocated that alum sludge could be a potential recyclable product, offering great commercial potential for reuse (Goldbold et al., 2003; Rensburg and Morgenthal, 2003). Therefore, with a continual increase in the production of waterworks sludge certain (at least for now), and compliance with the prevailing legislative and economic drives pointing toward waste avoidance and the beneficial reuse of waste streams necessary, a number of constructive attempts and research efforts have been made, particularly in recent years, to reuse alum sludge. These include laboratory and full-scale attempts to use alum sludge as a component in the manufacture of several materials, such as concrete or cement mortars (Goldbold et al., 2003); as geotechnical works materials (Carvalho and Antas, 2005); for use in agriculture and silviculture (Moodley and Hughes, 2005; Titshall and Hughes, 2005); as a primary source of Al-based coagulants through several recovery processes (Stendahl et al., 2005); and for P reduction during wastewater treatment (Kim et al., 2003). Fundamentally, these approaches for beneficial reuses offer two distinct advantages – in terms of (i) economic savings on overall treatment plant operation costs and (ii) environmental sustainability.

Four categories of alum sludge reuse were identified during the literature search conducted in this study: (i) use of alum sludges as building and construction materials such as brick making (Anderson et al., 2003; Goldbold et al., 2003); manufacture of cement and cementitious material (Kikuchi, 2001; Goldbold et al., 2003; Pan et al., 2004) and as materials in pavement and geotechnical works (Carvalho and Antas; 2005); (ii) the land application of alum sludges involving the controlled spreading of the sludge onto or into the surface layer of soil to stabilise, degrade, and immobilise the sludge constituents (Elliot and Dempsey, 1991); for agricultural soil improvement (Dayton and Basta, 2001; Owen, 2002; Moodley and Hughes, 2005; Pecku et al., 2005); as a soil buffer (Elliot and Dempsey, 1991; Wang et al., 1998; Rensburg and Morgenthal, 2003), and for nutrient reduction in laden soils and runoffs (Liu et al., 1997; Sharpley, 1997; Gallimore et al., 1999; Sampson and George, 2005); (iii) the use of alum sludges in wastewater treatment processes to enhance treatment performance in wastewater treatment processes, including its use as a coagulant in wastewater treatment (Horth et al. 1994; Wei, 1999; Chu, 2001); as an adsorbent for pollutants and metals immobilisation in wastewater treatment (Sujana et al., 1998; Huang and Chiswell, 2000; Zumpe et al., 2002; Kim et al., 2003; Wu et al., 2004a, 2004b; Georgantas and Grigoropoulou, 2005; Babatunde et al., 2005; Yang et al., 2006a, 2006b; Razali et al., 2007; Babatunde et al., 2009); and (iv) co-conditioning and dewatering with sewage sludge (Lin et al., 2001; Zhao, 2002; Lai and Liu, 2004; Yang et al., 2009).

With respect to using alum sludge to enhance P removal during wastewater treatment, the basic idea is that the abundant amorphous Al ions in alum sludge can become valuable for P removal in wastewater treatment, since such ions have been demonstrated to enhance the processes of adsorption and chemical precipitation that aid P immobilisation (Kim et al., 2003; de-Bashan and Bashan, 2004; Babatunde et al., 2005; Yang et al., 2006a, 2006b). The P-adsorption capacity of alum sludges reported in literature is summarised in Table 2.1. It is noted that alum sludge has an excellent P-adsorption capacity while pH plays a key role in the P-adsorption process. More recently, Makris et al. (2006a, 2006b) reported the use of alum sludge for other pollutant immobilisation, such as perchlorate and arsenic.

Table 2.1. Maximum P-adsorption capacity of alum sludges reported in the literature.

Test conditions	Maximum adsorption capacity Q0 (mg PO_4^{3-}/g sludge)			Reference
	Ortho-P	Poly-P	Organic-P	
Particle size <2.36 mm pH 4.0 Equilibration time 1 day	10.2 (Initial P concentration 14.7 mg PO_4^{3-}/L)	7.4 (Initial P concentration 10.8 mg PO_4^{3-}/L)	4.8 (Initial P concentration 3.3 mg PO_4^{3-}/L)	Razali et al., 2007
Particle size <2 mm Initial P concentration up to 4,000 mg PO_4^{3-}/L pH 7.1 Equilibration time overnight	25.0	14.3	12.5	Kim et al., 2003
Particle size 0.063 to 2.36 mm Initial P concentration 15.3 mg PO_4^{3-}/L pH 4.3	10.5	☐	☐	Yang et al., 2006a
NS*	0.30-0.33	☐	☐	Huang & Chiswell, 2000
Particle size 0.1 to 0.3 mm Initial P concentration 918 mg PO_4^{3-}/L Equilibration time 1 day	38.3	☐	☐	Ippolito et al., 2003
Particle size <2 mm Initial P concentration 10.7g PO_4^{3-}/L Equilibration time 17 hrs	2.02-50.49	☐	☐	Dayton et al., 2003
Particle size <150 µm Initial P concentration 0-10.7 g PO_4^{3-}/L	5.63-90.27 (Equilibration time 17 hrs) 31.82-113.22 (Equilibration time 6 days)	☐	☐	Dayton & Basta, 2005b
Particle size <2 mm Initial P loading 30.6 mg PO_4^{3-}/g sludge Equilibration time 10 days	22.95-30.60 (four kinds of alum sludge tested)	☐	☐	Makris and O'Connor, 2007

NS*: not specified.

2.2 Constructed Wetlands for Environmental Pollution Control with a Special Focus on Ireland

The last ten years in Ireland have seen the significant introduction of CW technology as a sewage-treatment system for single households and small rural communities and in some cases as a tertiary treatment stage for larger villages and towns. They have also gained increasing popularity in the treatment of agricultural wastewaters (Healy et al., 2007). In particular, another EPA-funded project, WETBALANCE, is focused on investigating the use of CW for the treatment of wastewater from a dairy-processing plant. Considerable synergy was explored with this project in that the concept of alum sludge being promoted in the current report can be used to enhance P removal in such wastewaters. Constructed wetlands have also been considered as alternative treatment systems in cases where a site is deemed unsuitable for septic tank construction (Gill et al., 2004). In addition, research interests in CW techniques have developed rapidly in recent years. Several authors, such as Haberl et al. (1998), Lienard et al. (1998), Greenway and Woolley (1999), Tanner et al., (2000), Vymazal (2002) and Rousseau et al. (2004) have published papers relating to the application of CW for environmental pollution control in their respective domains.

2.2.1 Current Status of Constructed Wetland Systems in Ireland

2.2.1.1 A Glance

According to an inventory survey carried out in 2005 at University College Dublin (UCD), there are now over 140 CW sites in Ireland (Babatunde et al., 2008a). Figure 2.1 illustrates the geographic distribution of these CW across the country. From Fig. 2.1, it is believed that CW have been established successfully in all parts of Ireland without any exception. There is no reported negative effect from the Irish climate on the performance of the CW.

Figure 2.1 indicates that the county with the highest number of CWs in Ireland is Wexford, followed by Cork. The two counties account for about 30% of all the CWs in Ireland. Both Wexford and Cork are predominantly rural areas. However, while 47% of the systems in Wexford are used for treating wastewater emanating from dwellings, approximately the same numbers of systems (44%) are used for commercial purposes in Cork. In the urban areas, it would seem that conventional wastewater treatment technologies are still dominant, for example in Co. Dublin, which has only about 2% of the total number of CW. Therefore, given the economic constraints of installing conventional wastewater treatment systems in most cases, CW would be a cost-effective land-use practice, which could help retain the nutrients and improve surface water quality.

Figure 2.1. Geographic distribution of constructed wetlands across Ireland.

2.2.1.2 *Types of Constructed Wetland Systems Used and Wastewater Treated*

The main types of CW and the types of wastewaters treated in Ireland are shown in Fig. 2.2. This shows that most CWs in Ireland are designed to treat municipal or domestic wastewater. However, some CWs treat domestic wastewater exclusively while others do so in combination with other sewage such as dairy washings, restaurants, etc. There is also an increasing trend of applying CW to farmyard waste management and urban storm water runoff (in particular, motorway runoff). Figure 2.2 highlights that surface flow (SF) and free water systems (FS) are the most used types of CW in Ireland.

2.2.1.3 *Treatment Performances*

Results from 13 case studies indicate an average removal of 76.8 to 99.8% BOD_5, 76.3 to 9.7% COD and 67 to 99.9% NH_4–N. Despite the wide range and variations of inlet concentrations of the pollutants, most CWs can be judged to have performed well. A limited comparison was made between these performances and those reported for other countries as compiled and presented in Babatunde et al. (2008a). At first, it seems conclusive that the Irish CWs have a superior performance when compared to CWs in other countries in Europe. It must however be noted that the removal efficiencies reported are based on inlet–outlet concentrations and the average inlet concentrations in most cases were quite high for the Irish CWs. This may lead to significantly biased results as it has been well established that removal efficiencies increase with increasing inlet concentrations (Vymazal, 2002).

2.2.1.4 *Special Case: The Irish Integrated Constructed Wetlands*

The Irish Integrated Constructed Wetlands (ICWs) are an initiative of Duchas, the Irish Heritage Service and they represent a specific design approach to CWs. The approach strives to achieve ‘landscape fit’ and ‘biodiversity’ into the design of CWs. Fundamental to their design is water quality improvement and the incorporation of the widest possible range of ecological conditions normally found in natural wetlands (including those of soil, water, plant and animal ecology) (Dunne et al., 2005a, 2005b). They are seen as a potential wastewater solution for agriculture, agribusiness, rural communities and the environment. As part of a Duchas-led ecological restoration project in the Anne valley on the south coast of Co. Waterford, Ireland,

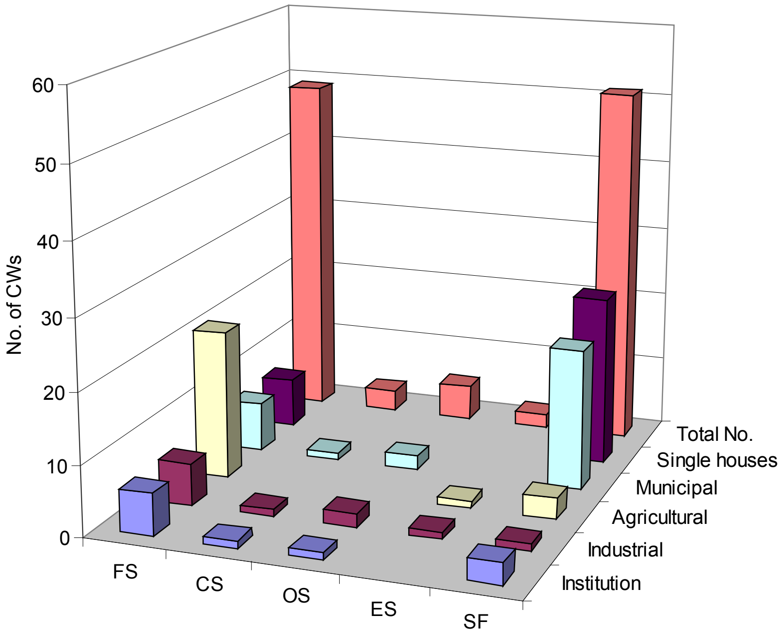


Figure 2.2. Types of constructed wetland and wastewater treated in Ireland: free water systems (FS); combined systems (CS); other systems such as sludge drying systems and stream filtration basins (OS); experimental systems (ES); surface flow systems (SF).

more than 13 ICWs were constructed to enhance water quality management and prevent environmental pollution within the Anne valley. It is reported that within a year of the ICW commissioning, approximately 75% of farmyard runoff was intercepted, leading to improvements in the receiving surface waters of the catchment. Details of the ICW design and construction are published in Dunne et al. (2005a, 2005b) and Harrington and McInnes (2009).

2.2.2 *Constructed Wetland Research in Ireland*

2.2.2.1 Research Output

An evaluation of progress made so far in CW research in Ireland was carried out. Prior to this evaluation, a comparative analysis had been carried out in 2007 to assess research output on CW in Ireland in comparison to other countries. The 2007 analysis was focused on research outputs and on the numbers of papers published in over 67 journal sources, and papers selected for publication from presentations in the bi-annual international conference on Wetland Systems for Water Pollution Control. The numbers of published papers were analysed statistically using the Web of Science performance indicator charts. The performance indicator charts had a database search output of 67 journal sources, covering 1107 CW papers published between 1992 and 2002 worldwide. Selected papers from presentations made at the biannual conference between 1992 and 2004 were also analysed. The results obtained from the analysis, which are published in Babatunde et al. (2008a), show that one of the main findings is that there has been very little published of either research findings or of treatment performances of operating CWs in Ireland.

A similar comparative analysis was carried out in 2010. However, the period of coverage was 2003 to 2009 for published papers; for papers selected for publication at the biannual international conference on CW for water pollution control, updated data were collated from the 2006 and 2008 conferences. A total of 1633 published papers on CWs between 2003 and 2009 were analysed in addition to the selected presentations from the 2006 and 2008 biannual conferences. The previous and updated results are jointly shown in Table

Table 2.2. Research outputs on constructed wetlands in selected countries.

Country	[a] No. of journal papers	[b] No. of journal papers	Conference presentations					
			Cumulative papers		Author affiliation			
			A	b	Academic		Collaboration	
					a	b	a	b
USA	551	515	68	12	29	7	13	5
UK	96	94	34	8	23	3	2	1
Australia	70	65	41	10	19	4	5	5
Germany	56	92	18	9	12	4	3	3
Canada	45	87	6	3	3	2	1	1
Sweden	30	49	8	0	8	0	0	0
New Zealand	28	20	14	2	8	0	2	1
Norway	23	34	10	0	6	0	3	0
Spain	20	85	3	8	3	6	0	2
Netherlands	17	19	2	2	2	1	0	1
Austria	14	28	11	4	6	2	0	1
France	14	39	8	7	5	4	0	3
Denmark	11	20	11	1	8	1	2	0
Belgium	4	20	4	2	2	2	2	0
Ireland	3	40	1	2	1	2	0	0
Slovenia	2	4	4	0	2	0	2	0

a refers to the period 1992–2002.

b refers to the period 2003–2009.

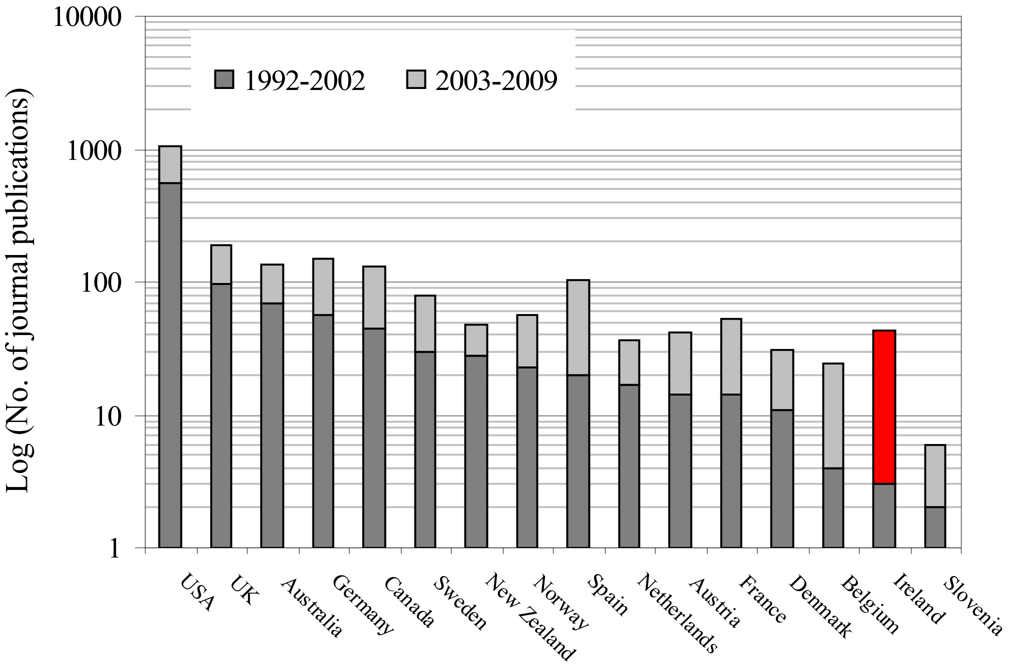


Figure 2.3. Number of published articles on constructed wetlands in selected countries during the periods 1999 to 2002 and 2003 to 2009.

2.2 while a plot presenting the data on the number of published papers from the two analyses is shown in Fig. 2.3. From Table 2.2, it can be seen that the leading countries in CW still maintain their lead in terms of published papers on research work; these include the USA, UK, Australia and Germany. These four countries also accounted for 56% of papers selected for publication from the biannual CW conference for 2006 and 2008. What is also worth noting is that 50 to 100% of the research carried out in these four countries has been a collaboration between university and industry (i.e. 2003 to 2009).

However, it is interesting to note that Ireland had the most significant jump in the number of publications on CW between the first and second analysis (see Fig. 2.3). The number of published articles on CW in Ireland increased from 3 (1992 to 2002) to 40 (2003 to 2009). This underlines that CWs and CW research are now gaining more attention in Ireland. Although Ireland has the 14th lowest population among the 16 selected countries that were analysed, it had the greatest increase in research outputs between the two periods – ranking 8 out of 16 (second period) compared with a ranking of 15 out of 16 (first period) based on the number of publications. However, attendance and presentation at conferences by researchers in Ireland working in the CW field is still an area that needs improvement. Furthermore, although the dataset from publications selected from the bi-annual conference is quite small, it would appear that CW research in Ireland is still university led with very little industrial collaboration. It is therefore suggested that a more significant partnership between university and industry needs to be forged to further the development of CW in Ireland.

3 Optimising the Configuration of Alum Sludge-Based Constructed Wetlands

Alum sludge has been shown to have an excellent P-adsorption ability and has been proposed to be used as a substrate in CWs for wastewater treatment. The study in this period was aimed at assessing and determining the optimal configuration needed for an alum sludge-based CW system using six laboratory-scale CW reactors. The reactors were configured using alum sludge as the main substrate with or without gravel/sand at the infiltrative surface. The optimal configuration was set to be the configuration that gave the best pollutant removal performance without other side effects (such as clogging).

3.1 Materials and Methods

3.1.1 Alum Sludge

Dewatered alum sludge cakes were collected from the dewatering unit of the Ballymore-Eustace Water Treatment Plant located in Co. Kildare (Fig. 3.1). The plant uses a nearby Poulaphuca reservoir water to produce 230,000 m^3/d of potable water for Dublin city. The alum sludge produced in the plant is a brownish-green residual derived from the treatment of a medium colour, medium turbidity, raw water that is sourced from the Dublin and Wicklow mountains, which are upland catchments of peat over granite bedrock. Using the traditional processes of flocculation, sedimentation, filtration and disinfection, the plant uses aluminium sulphate as a coagulant (with a dosage of 40 to 65 mg/L) to treat the source water. Currently, alum sludge from the sedimentation tank together with the backwashing stream from the filtration tank is thickened and then conditioned with an organic polymer (Magnafloc LT25) at a dosage of 0.6 to 1.5 mg/L and finally dewatered by filter press to produce sludge cakes for landfill as a final disposal.

The collected alum sludge cakes (with solids content of 23%) were air dried and then ground and sieved to prepare the samples for testing. Characterisation of the sludges was conducted using energy dispersive spectrometry (EDS). The major elemental components, including Al, Fe, Ca, Mg, P Si, Cu, Zn, Cd and Pb, as well as humic acids classified as total organic carbon (TOC) of the alum sludges were determined by inductively coupled plasma optical emission spectrum (ICP-OES, IRIS Intrepid II XSP), inductively coupled plasma mass spectrometry (ICP-MS), Fourier transform infrared (FT-IR) Spectrophotometry (EQUINOX-55) and total organic carbon analysis (high sensitivity) TOC-V CSH (from Shimadzu). The energy dispersive spectrum of the sludge is shown in Fig. 3.2. The chemical components have been identified clearly and Table 3.1 lists them in mass percentage.

Table 3.1. Chemical components of dewatered alum sludge (DAS).

Chemical composition	Unit	Ballymore-Eustace sludge
Al	mg/g DAS	169.52
P	mg/g DAS	0.582
Ca	mg/g DAS	4.03
Fe	mg/g DAS	6.65
Mg	mg/g DAS	0.251
Pb	mg/g DAS	0.019
Cd	µg /g DAS	0.51
Zn	mg/g DAS	0.065
Cu	mg/g DAS	0.069
Si	mg/g DAS	3.11
TOC	mg-C/g DAS	172.6

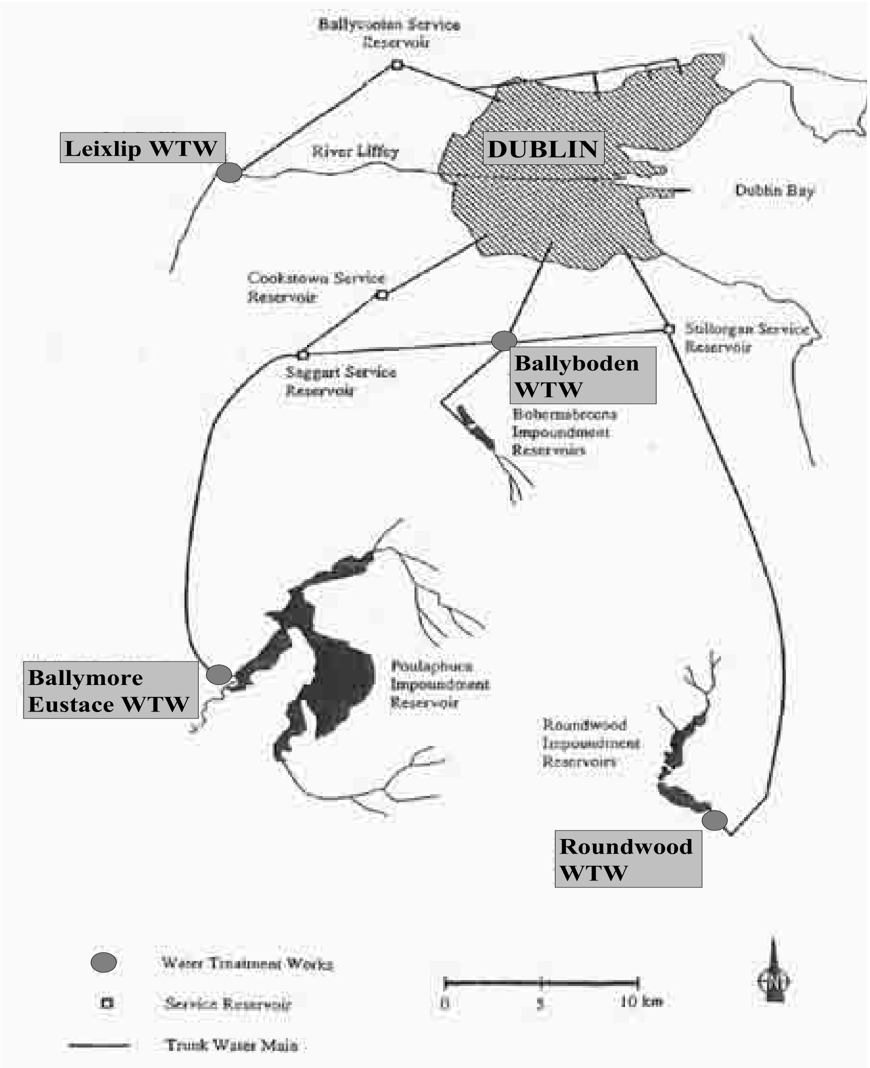
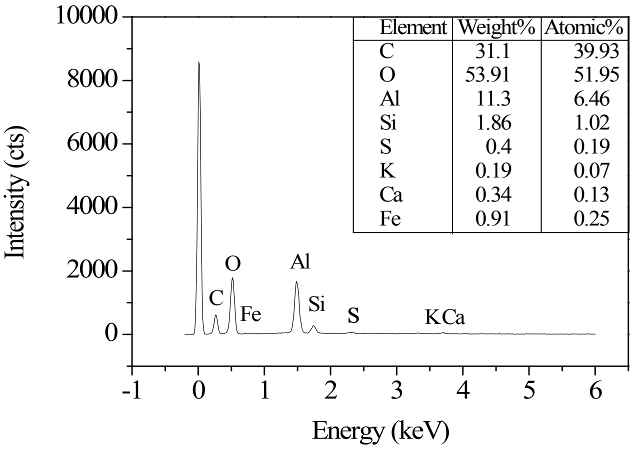


Figure 3.1. Geographic layout of water-treatment works (WTW) in Dublin.



Element	Weight%	Atomic%
C	31.1	39.93
O	53.91	51.95
Al	11.3	6.46
Si	1.86	1.02
S	0.4	0.19
K	0.19	0.07
Ca	0.34	0.13
Fe	0.91	0.25

Figure 3.2. Energy dispersive spectrum of the alum sludge collected from Ballymore-Eustace water treatment plant.

3.1.2 CW Reactor Set-up

Six different vertical flow CW (VFCW) reactors were constructed using Pyrex columns (1.0 m long, Ø = 0.095 m). Figure 3.3 shows a photographic description of the reactors while the detailed design and operation of the reactors is given in Table 3.2. The Pyrex columns were filled with the dewatered alum sludge and overlain with either pea gravel or sand in different proportions, ranging from 0 to 60%.

Figure 3.3. Laboratory-scale vertical flow constructed wetland reactors filled with different proportions of dewatered alum sludge.

Table 3.2. Composition and design of the vertical flow constructed wetland reactors.

	Constructed wetland reactors					
Composition (%)	**1**	**2**	**3**	**4**	**5**	**6**
Alum sludge	100	80	60	40	60	100
Pea gravel	0	20	40	60	20	0
Sand	0	0	0	0	20	0
^{a} Layout (cm)						
Sharp sand	0	0	0	0	10	0
Pea gravel	0	10	20	30	10	10
Sludge	50	40	30	20	30	50
Gravel support	10	10	10	10	10	0
Design						
Flow type	Batch	Batch	Batch	Batch	Batch	Continuous
No. of daily cycles	6–8	6–8	6–8	6–8	6–8	–
^{b} Pore volume (l)	1.94	1.74	1.63	1.55	1.89	1.8
^{c} HLR (m^3/m^2.d)	1.86	1.75	1.69	1.63	1.69	1.71
Flow rate (ml/min)	66	66	66	66	66	7.2
^{d} Contact time (hrs)	3	3	3	3	3	3

a in the direction of flow, b based on initial saturated pore volume, c hydraulic loading rate (HLR), d calculated based on 8 cycles/day.
d. = day

3.1.3 CW Reactors Operation

The reactors were fed from the same feed tank with the supernatant of a real agricultural farm wastewater (Table 3.3). During the operation, the supernatant was sometimes diluted with laboratory distilled water or spiked with potassium dihydrogen phosphate (KH_2PO_4) to achieve the desired operational P concentrations. Prior to being operated, the unsaturated pore volumes of the reactors were noted. The hydraulic loading rate (HLR) design was, however, based on the saturated pore volumes in each of the reactors. The HLR varied slightly across the reactors owing to differences in their saturated pore volumes, which is caused by the difference in their configurations. The reactor with the 100% dewatered alum sludge had the highest pore volume, meaning that a comparatively higher volume of wastewater could be treated, and with a corresponding higher loading.

Table 3.3. Mean concentrations of wastewater parameters fed into the constructed wetland reactors.

	Operation period				
	1	**2**	**3**	**4**	**5**
Parameter	Mean	Mean	Mean	Mean	Mean
COD (mg/L)	214.4	399.4	399.4	213.6	1230.7
SS (mg/L)	85.0	97.2	170.3	231.5	100.2
RP (mg-P/L)	18.1	35.3	53.2	207.3	346.1
SRP (mg P/L)	9.8	19.0	34.2	172.3	309.9

Period 1: 12/06/06 to 02/07/06, Period 2: 10/07/06 to 30/07/06, Period 3: 07/08/06 to 27/08/06, Period 4: 04/09/06 to 24/09/06, Period 5: 16/10/06 to 22/01/07, Period 5 includes period of closure due to holidays, SD = standard deviation; SS = suspended solids; RP = reactive phosphorus; SRP = soluble reactive phosphorus.

The pumping operation was conducted in batches using peristaltic pumps. Experiments were conducted over five P loading periods, and each corresponds to a specific P loading and operating condition. The total duration of the experiments was 25 weeks. Reactor 6 was operated in a continuous up-flow mode to examine any effect of the batch feeding on P removal (in comparison to Reactor 1) and also any reduction in clogging tendency by avoiding surface filtration of the suspended solids (SS). It should however be noted that the upward flow may be potentially more expensive, and its benefits should be weighed against its cost.

3.1.4 CW Reactor Monitoring

The performance of each reactor was assessed by chemical analysis of the inflow and outflow samples and visual inspection in order to determine the optimal performing configuration. Samples were analysed for reactive phosphorus (RP, determined on unfiltered samples), soluble reactive phosphorus (SRP, determined on filtered samples), SS and COD. All analysis was done according to standard methods (APHA-AWWA-WEF, 1998). During the fifth period, the redox potential (Eh) in the reactors was measured using a portable redox meter. The Eh meter and electrode were tested prior to each use against solutions of excess quinhydrone in pH buffers. However, the Eh values were not pH corrected since the reactors were close to neutral pH.

3.2 Results

3.2.1 RP and SRP Removal

A clear influence of the proportion of dewatered alum sludge in the CW reactors on RP and SRP removal can be observed from the pattern of the removal efficiency obtained. During the first period, all the reactors were able to remove significant amounts of RP and SRP, albeit at slightly different efficiencies. However, beyond this period, differences soon began to emerge, particularly in Reactor 4, which had the least proportion of alum sludge. Over time, the decline in P removal in Reactor 4 was steady, whereas the other reactors still had a high P removal efficiency which was very comparable between these reactors. At the end of the experimental period, an average overall P removal efficiency of 93.1, 91.9, 79.5, 67.1, 89.8 and 86.9% (SRP), and 87.3, 87.8, 77.0, 69.0, 86.4 and 83.6% (RP) was achieved in Reactors 1, 2, 3, 4, 5 and 6 respectively. This clearly reflects the beneficial advantage of the inclusion of alum sludge in the reactors. Comparatively, Reactor 4, which had the highest proportion of pea gravel and the least proportion of alum sludge, had the lowest P removal efficiency. Obviously, this was because it had the least proportion (40%) of alum sludge in its composition, and since the pea gravel is not likely to contribute significantly to P removal in the system, the alum sludge must have been significantly loaded, resulting in the decline in its performance and overall poor removal efficiency of the reactor.

3.2.2 SS Removal

The plot of SS removal over time and the statistical performance analysis are presented in Fig. 3.4. The figure shows that the fluctuation trend in the inflow is mirrored to some extent in the outflow SS concentrations from the reactors. This indicates at least some dependence of the level of SS in the effluent quality on the level of SS in the influent, as peaks in the influent mirrored peaks in the effluent. Although Reactor 5 had the lowest mean effluent outflow quality at the end of the experimental period, there was very little difference between the performances of the top three performing reactors as regards SS removal (i.e. Reactors 1, 2 and 5).

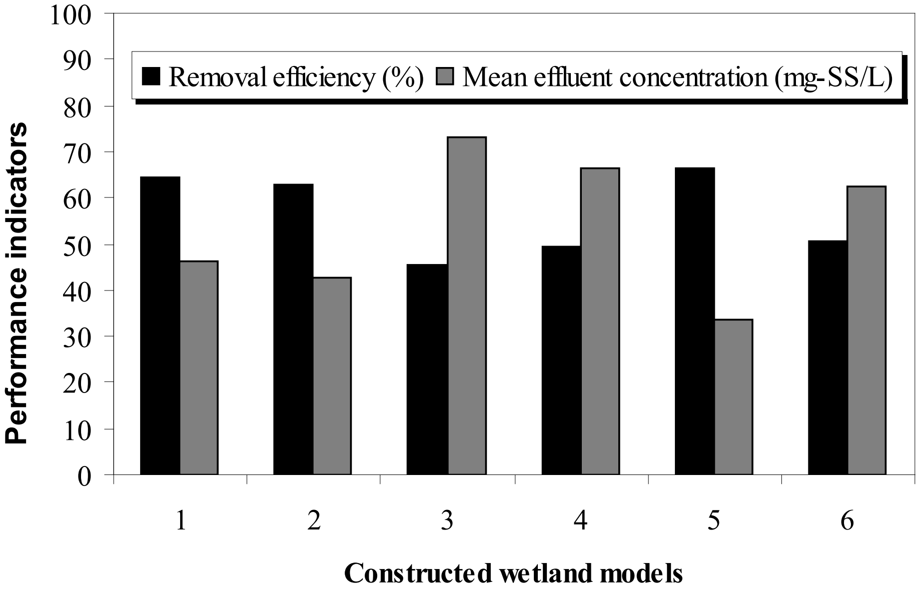


Figure 3.4. Comparative mean effluent concentration and overall removal efficiency of suspended solids (SS) for the constructed wetland reactors.

Surprisingly, Reactor 6 did not present any superior SS removal, particularly in comparison to Reactor 1. Frequently, the effluent from Reactor 6 appeared more turbid, suggesting that either the upward flow of water had an uplift effect on suspended particles in the bed or, since the flow is continuous, solids in the influent are not able to fully settle/deposit on the alum sludge grain, or even that, when settled, they are subsequently resuspended. Reactor 5 had an average overall removal efficiencies of 66.4%, whereas Reactors 1 and 2 had average overall removal efficiencies of 64.6 and 63.0% respectively, giving a marginal difference of 1.8 to 3.4% in the removal efficiency. The inclusion of pea gravel at the infiltrative surface of the reactors beyond 10 cm (in the direction of flow) did not prove to have any beneficial advantage.

3.2.3 COD Removal

All reactors were assessed for COD removal (Table 3.4). An initial COD removal ranging from 27.0 to 64.0% (see Period 1 in Table 3.4) was obtained and this corresponds to a mean SS removal of 75.1 to 88.3%. At this stage, the removal of SS and COD can be described as being positively related. However, beyond the second period and up to the fourth period, there was a steady decline in COD removal across all the reactors, resulting in a trend of negative COD removal. One explanation for this phenomenon might be that the reactors may be acting as a source rather than as a sink for removing organic matter. During this same period, there was only a marginal decrease in SS removal across the reactors. The COD in the effluent must have been caused by other phenomena. It is thus likely that the sustained removal of P at very high inlet P concentration may have initiated the release of some initially adsorbed organics into the effluent.

Analyses of redox potential values (see Table 3.5) indicated that all the reactors were transiting from oxidising to reducing conditions, and they were all generally in moderately reducing conditions (Eh between +100 and +300 mV). Consequently, an anaerobic condition is likely to have prevailed across all the reactors at the time the readings were taken. Although the Eh is a semi-quantitative measure, it can be seen from Table 3.5 that Reactor 6 was in the most reducing condition (Eh ≈ +100 mV), while Reactor 1 was in the least reducing condition of all the reactors (Eh = 288 mV). This gives an indirect assessment of the beneficial effect of the intermittent strategy in promoting aerobic/oxidising conditions in the reactors. The dissolved oxygen (DO) values also indicate very low oxygen levels. These conditions reflect both persistent low oxygen and limited aerobic biodegradability in the reactors. There was also a mild olfactory detection of hydrogen sulphide gas and this also lends credence to the anaerobic state of the (CW) reactors. Therefore, it might be reasonable

Table 3.4. COD removal performance in the six constructed wetland reactors.

	Constructed wetland reactors					
	1	**2**	**3**	**4**	**5**	**6**
Period 1						
Mean	116.2	116.0	118.8	132.8	73.6	153.2
SD	27.3	257.2	15.8	29.5	42.5	22.7
% Removal	44.1	46.1	43.5	36.3	64.0	27.0
Period 2						
Mean	262.2	231.2	267.4	263.6	212.9	291.4
SD	83.2	70.9	62.5	61.4	58.5	62.7
% Removal	22.7	41.5	20.3	33.1	46.0	25.7
Period 3						
Mean	298.7	251.1	289.8	323.8	277.3	390.8
SD	95.4	62.6	53.4	43.9	77.3	62.2
% Removal	7.1	20.4	8.5	-2.4	11.0	-24.7
Period 4						
Mean	449.2	448.5	434.8	509.2	466.9	462.9
SD	46.9	39.1	39.4	54.8	68.9	89.4
% Removal	-137.2	-138.9	-132.3	-175.5	-154.5	-134.7
Period 5						
Mean	820.5	879.9	900.0	838.1	841.0	954.7
SD	204.3	206.1	211.8	203.4	177.2	288.7
% Removal	24.8	20.9	19.2	23.2	17.6	18.1

SD = standard deviation.

to suggest that anaerobic degradation is prevailing in the reactors. As the degradation action of anaerobic bacteria is much slower than with aerobic bacteria, there is a possibility of incomplete utilisation of the dissolved/hydrolysed organic matter. This may have contributed to the high COD in the effluent and the negative COD removal efficiency. Consequently, 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 induce a release of initially adsorbed organics. This finding is beneficial to understanding the removal mechanism and to ensuring that CW operators should be aware of the possibility.

3.2.4 Optimal Configuration

The optimal configuration of the CW reactor is taken as that which gives the best treatment performance in terms of pollutant removal and which does not clog easily. In terms of P removal, the performance of the

Table 3.5. Selected and monitored parameters in the constructed wetland reactors.

	Constructed wetland reactor					
Parameter	1	2	3	4	5	6
[a]Eh (mV)	+288	+255	+209	+258	+209	+119.5
DO (mg/L)	1	0.3	0.7	0.4	0.4	0.5
pH	7.22	6.96	6.87	6.81	6.81	7.04
[b]H2S	Y	Y	Y	Y	Y	Y

[a] mean value of three sets of readings [b] based on olfactory detection, Y = positive detection. All data were taken during the fifth period.

systems decreased with an increase in the proportion of gravel used in the substrate configuration. Consequently, the optimal system configuration for P removal would be the system configured with 100% dewatered alum sludge in its substrate. In the case of SS removal, the inclusion of gravel at the infiltrative surface of the reactors appears to have marginally increased the solids storage capacity of the systems, but, nonetheless, there was no significant difference in SS removal in the systems based on the use of gravel at their infiltrative surface. In addition, all the systems exhibited clogging tendencies. As far as clogging is concerned, the optimal beneficial configuration would appear to be that in which the use of gravel at the infiltrative surface is limited to the 0 to 10 cm range. Initial COD removal was attributed to SS removal and this is also linked to the efficiency of the reactors at removing SS. However, the high influent P loading appears to have impaired COD removal efficiencies in the reactors. Consequently, the optimal configuration for the alum sludge-based CW appears to be that in which the use of gravel at the infiltrative surface (if used at all) is limited to the top 0 to 10 cm.

3.3 Summary

The main results derived from this study have shown that all the reactors exhibited clogging tendencies, irrespective of the substrate configuration or the infiltrative surface layer. There was no difference in the efficiency of SS removal among the reactors, indicating that there was no significant benefit derived in using gravel at the infiltrative surface, particularly beyond the 0 to 10 cm range. However, the reactors were efficient in removing P even at higher loadings, but the P removal efficiency decreased with the increase in the proportion of gravel in the substrate configuration. Initial COD results show excellent removal. However, the preferential adsorption of P onto the surface of the alum sludge at high influent P loading appears to have initiated a release of initially adsorbed and un-degraded organics. Consequently, it is inferred that although competition from other negative ions and biofilm development did not have any negative effect on P removal, a high P concentration may induce a release of initially adsorbed and un-degraded organics. Overall, results suggest that the optimal configuration would be that in which the use of gravel at the infiltrative surface (if used at all) is limited to the topmost 0 to 10 cm.

4 Long-Term Trial of a Single Alum Sludge-Based Constructed Wetland

The objective of the study in this stage of development was to examine, in controlled laboratory experiments, the effectiveness of a model CW with dewatered alum sludge as a main filter medium treating a real P-rich wastewater under a long-term trial. It is expected that the study will serve as a primer towards the establishment of an alum sludge-based CW system for real animal farm wastewater treatment.

4.1 Experimental Set-up

4.1.1 Alum Sludge

Dewatered alum sludge was collected from Ballymore-Eustace Water Treatment Plant (described in detail in Section 3.1.1).

4.1.2 Model CW Set-up and Operation

The single model alum sludge-based CW under investigation was set up using a 145 mm (internal diameter) Pyrex column filled firstly with gravel to 10 cm as a support layer followed by 35 cm in depth of prepared alum sludge (2.5 kg) as the main substrate. *Phragmites australis* (common reed) was planted on the top of the CW. The supernatant of the collected wastewater (with or without dilution with tap water) from an animal farm which includes about 2000 livestock units of sheep, pigs, cattle and horses was used as the influent with concentrations of 213±127 mg/L (COD), 110±69 mg/L (BOD_5), 28±15 mg/L (PO_4^{3-}-P) and 72±66 mg/L (SS), at a pH of 6.8±0.4. The influent was loaded onto the CW via a peristaltic pump from a feed tank at a daily flow rate of 8 L, which gives a hydraulic loading rate of 0.5 m^3/m^2.d (day (d.)). The system was operated using the tidal flow strategy for over 730 days. The rhythmical filling and draining generated the tides and this was realised using peristaltic pumps controlled by a preset electronic timer. The laboratory set-up of the model alum sludge-based tidal flow CW system is shown in Fig. 4.1.

4.1.3 Analysis

During the testing period, samples of influent and effluent were collected and analysed periodically for BOD_5, COD, SS, P and pH (WTW pH meter, model 325, Germany) according to standard methods. P analysis was conducted in two parts: (i) samples were reacted directly (without filtration) with the reagent and analysed to determine the RP and (ii) samples were filtered using a 0.45 μm membrane filter and the filtrate was analysed to determine the SRP.

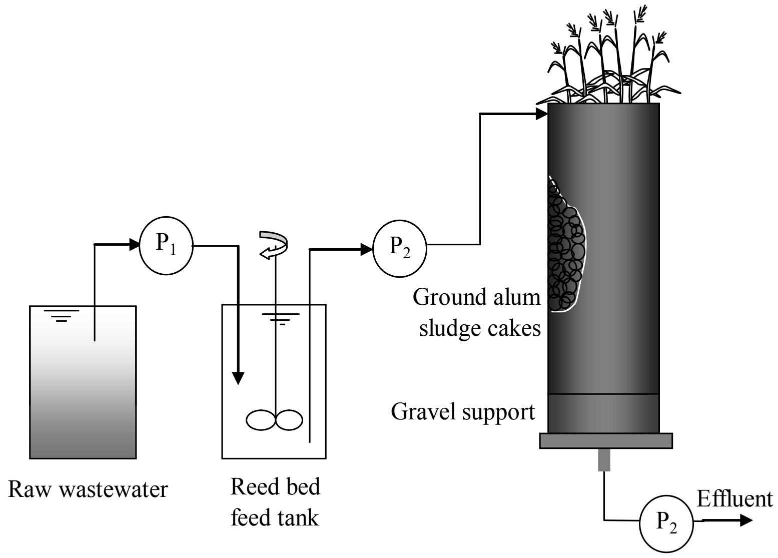


Figure 4.1. Laboratory set-up of the model alum sludge-based tidal flow constructed wetland system.

4.2 Results

Figure 4.2 illustrates the progressive treatment performance of the model CW. From the results, the initial removal of the carbonaceous substrates averaged 70% for BOD_5 and 56% for COD, although this would mainly be attributable to filtration because of the lack of biological activities in the newly set-up CW. However, with the gradual establishment of a dynamic biological system through intense activity of the reeds and microorganisms in the alum sludge matrix, improved removal of over 80% for both BOD_5 and COD was observed after 100 days' operation. Additionally, enhanced oxygen supply caused by the tidal flow operation strategy led to over 90% BOD_5 removal by the 260th day with a short period of exception as shown in Fig. 4.2. The calculated value of the theoretical oxygen supply rate by the tidal flow operation strategy used in the system is 137.2 $g/m^2.d$. An average removal of 73.3±15.9% for COD and 82.9±12.3% for BOD_5 was achieved during the entire experimental period. It can be seen from Fig. 4.2 that the COD removal exhibited some considerable fluctuations, and this may be caused by the integrated removal of enhanced adsorption, filtration and biological degradation of pollutants in terms of COD. Relatively, BOD_5 removal was solely from the biological degradation. The relative low removal efficiencies of BOD_5 and COD of the CW in the later stage of operation can be related to operational reasons as the laboratory was relocated during that period and this affected the routine operation.

Figure 4.3 provides evidence of a good correlation between BOD_5 loading and its removal (in $g/m^2.d$). Increased BOD_5 loading resulted in higher BOD_5 removal in terms of $g/m^2.d$ of the CW, indicating the intensive activities of the microorganisms inside the CW.

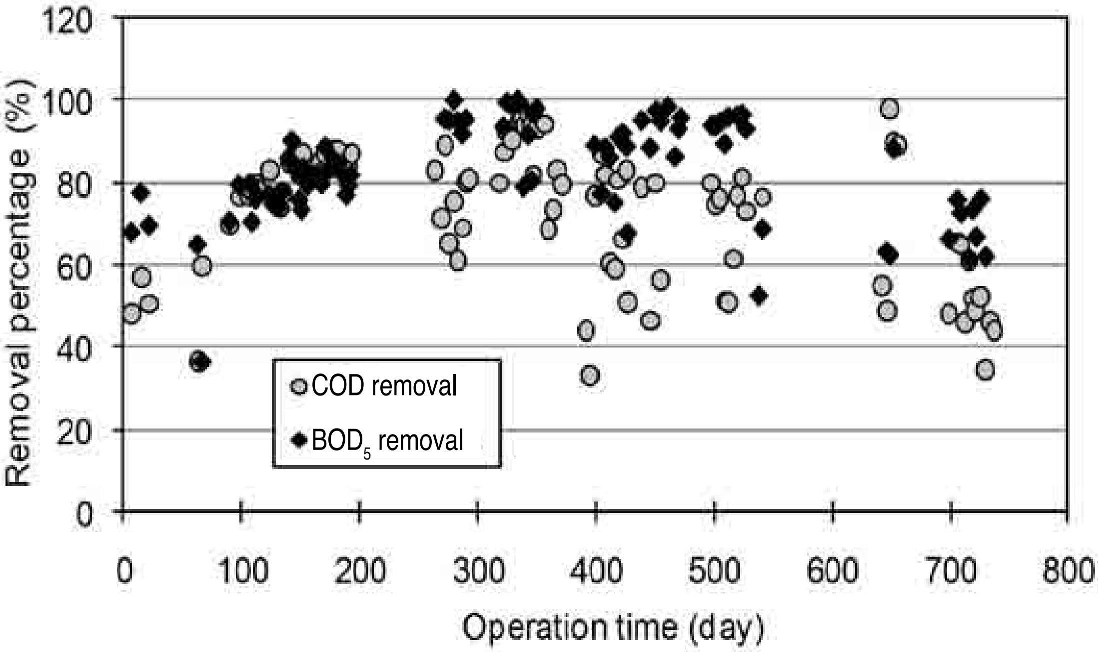


Figure 4.2. Removal efficiencies of COD and BOD_5 throughout the experimental period.

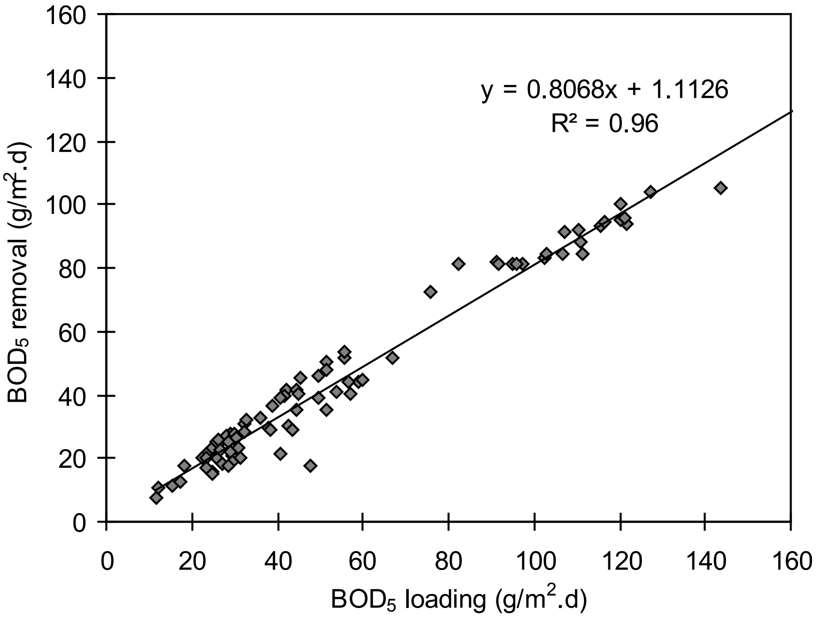


Figure 4.3. Correlation between BOD_5 loading and its removal.

Removal of both RP and SRP was significant in the CW throughout the experiments. Average removal efficiencies of 86.4±6.0% for RP and 88.6±7.2% for SRP, respectively, were achieved under P loading of 13.5±7.2 g-PO_4^{3-}/m^2.d for RP and 8.2±3.2 g-PO_4^{3-}/m^2.d for SRP respectively. A mass balance estimation shows that about 35.8 mg-PO_4^{3-}/g-sludge was adsorbed by the sludge in the CW during the operation. Interestingly, the sludge in the CW did not get to saturation point and a P removal efficiency of 70% was still obtained, although an obvious decline of removal efficiency is observed from Fig. 4.4.

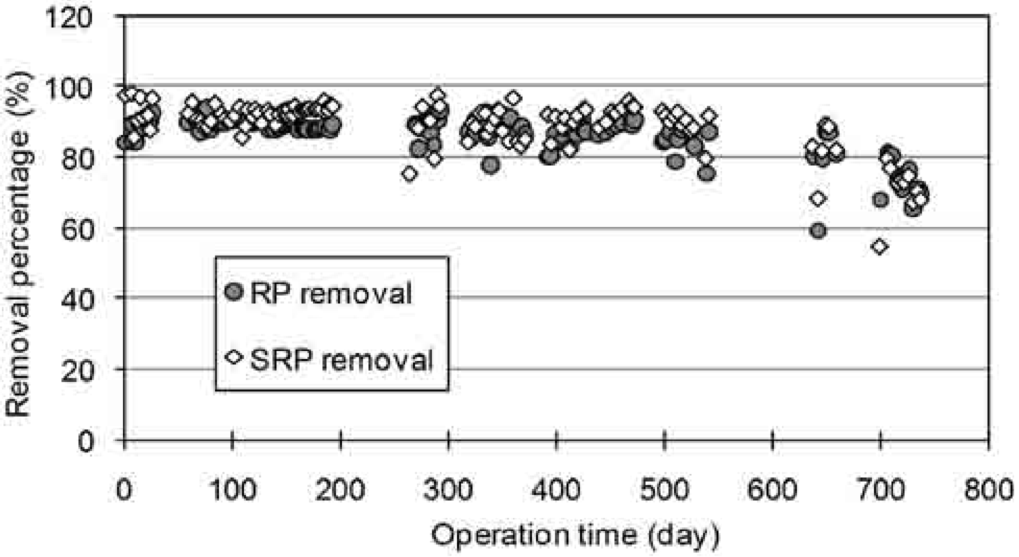


Figure 4.4. Removal efficiencies of reactive phosphorus (RP) and soluble reactive phosphorus (SRP) throughout the experimental period.

The trend of SS removal is shown in Fig. 4.5. Although an average SS removal of 77.6±17.5% was obtained, there was a significant fluctuation in the SS removal, and therefore further study of this issue may be warranted. However, ponding did not occur but concerns of CW clogging may warrant further investigation.

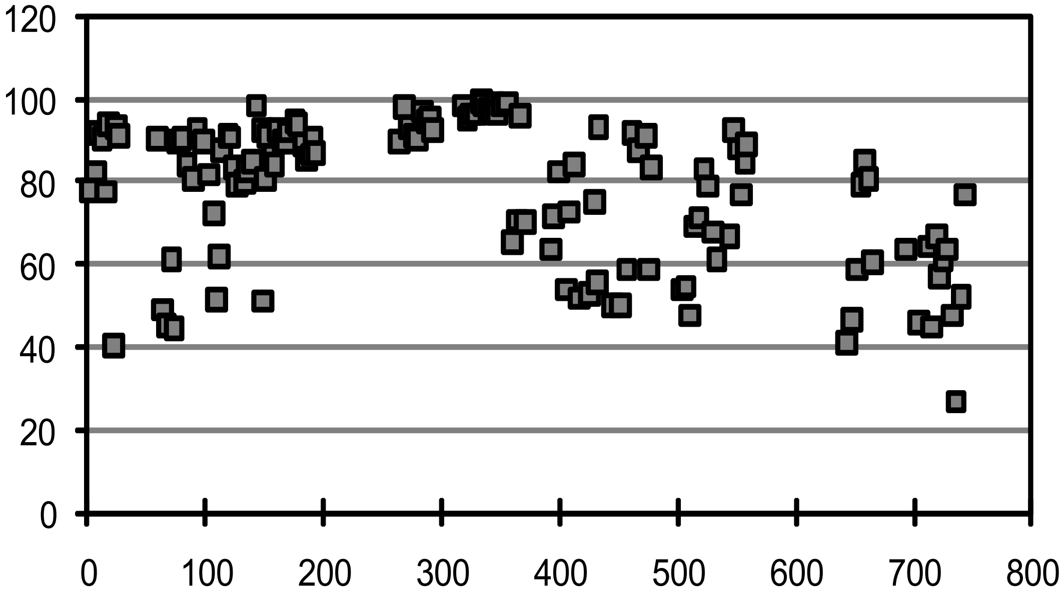


Figure 4.5. Removal efficiencies of suspended solids throughout the experimental period.

4.3 Discussion

4.3.1 BOD_5 and COD Removal

The goal of this period of study was to conduct a long-term examination of the alum sludge which is expected to act as a carrier for developing biofilm and also serve as adsorbent to enhance P immobilisation from treatment of a real wastewater. As a biofilm carrier, the sludge has demonstrated its role successfully. The significant removal of both BOD_5 and COD (see Fig. 4.2) have demonstrated the development of biofilm and intensive biological activity inside the CW. The results indicate that the model CW degraded the organics through biological activity inside the CW. A continuous reduction of COD and BOD_5 provides a definite indication of biological degradation of the organics from the wastewater.

4.3.2 Phosphorus Removal and the Lifetime of the Alum Sludge in the CW

During the monitoring period, the alum sludge exhibited good ability as an adsorbent to achieve high P immobilisation (see Fig. 4.4). Both RP and SRP were monitored in this study and there was a good correlation between them as illustrated in Fig. 4.6, suggesting that either RP or SRP can be used as a sole index for P removal in practice. Figure 4.7 shows the RP and SRP removal (in g/m^2.d) with the RP loading rate applied. It is noted that although the RP loading rates were varied from 6.2 to 32.2 g PO_4^{3-}-P/m^2.d, the CW system could remove P efficiently even as the loading rates increased, as indicated by a linear relationship between P loading and removal. From the linear regression analysis (see Fig. 4.7), it is interesting to note that the slopes of the two linear regressions are obviously different. If the removal of SRP is achieved mainly by adsorption and its biological assimilation by the growth of micoorganisms in the biofilm, then the difference between the two lines could be considered as an indication of the sludge's 'other' functions such as filtration and sedimentation. Obviously, adsorption is responsible for 42% (0.38/0.90) of P removal, based on the data of the current study. Thus, it is reasonable to state that the maximum adsorption capacity obtained experimentally from batch isotherm tests cannot be used as a guide to the actual lifetime of such CWs in real operation, although attempts have been made to do so in literature with little or no success (Arias and Brix, 2005; Dong et al., 2005; Park and Polprasert, 2008; Zhao et al., 2008a).

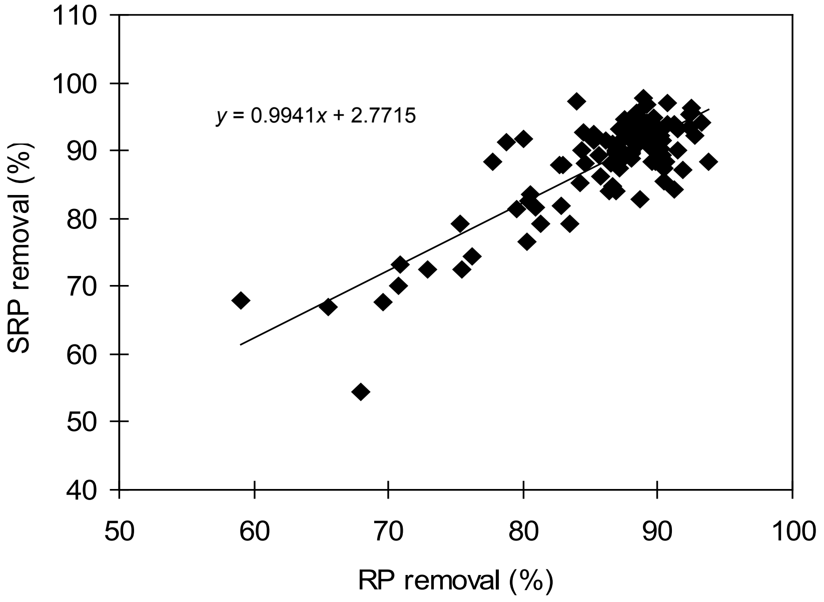


Figure 4.6. Correlation between reactive phosphorus (RP) and soluble reactive phosphorus (SRP) removal.

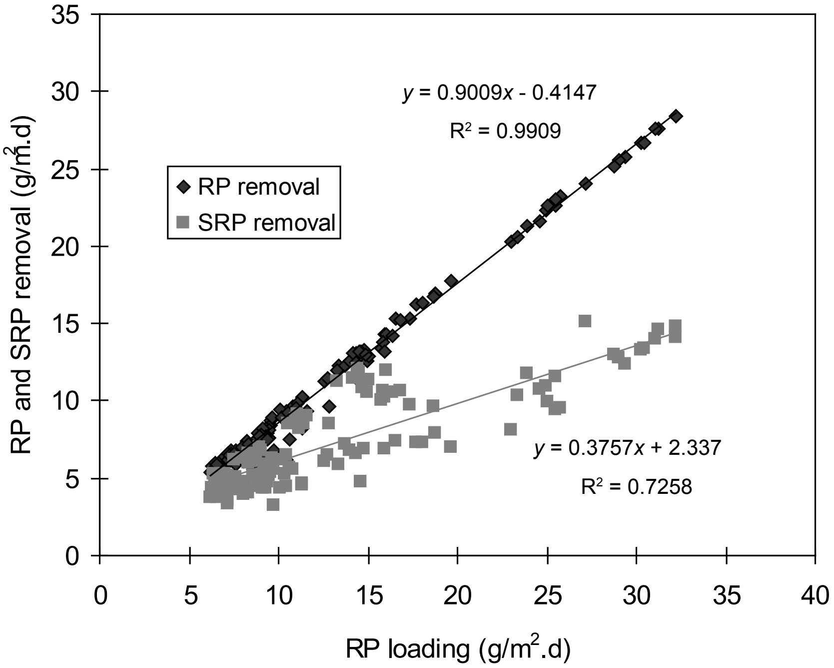


Figure 4.7. Correlation between reactive phosphorus (RP) loading and the removal (g/m^2.d) of RP and soluble reactive phosphorus (SRP).

4.4 Summary

The two-year experiment was conducted in a model alum sludge-based CW aimed at examining the feasibility and effectiveness of the CW in treating P-rich wastewater. Average removal efficiencies of 73.3±15.9% for COD, 82.9±12.3% for BOD_5, 86.4±6.0% for RP, 88.6±7.2% for SRP and 77.6±17.5% for SS were achieved during the entire operation period. The results have demonstrated that the dewatered alum sludge can be a good P adsorbent and a good carrier for biofilm attachment and growth. Results revealed that the 'P-adsorption proportion' by the sludge in the CW is 42% of overall P removal. Therefore, the lifetime of the alum sludge in the CW is reasonably longer than that determined from batch isotherm tests. Regarding the large-scale application of the CW system, further studies on clogging tendency and the possible release of substances inside the alum sludge are recommended.

5 Development of an Alum Sludge-Based Constructed Wetland System: Multistage System Trial

Using the single model alum sludge-based CW trial as a primer, the study described in this section is concerned with the design and performance analysis of a multistage model of the CW, which consists of four identical wetland cells connected in series using dewatered alum sludge as a main substrate for high-strength wastewater treatment. High-strength wastewaters are characterised as having an elevated concentration of organic matter and solids, or fats, oils and greases. They can be generated in any type of facility but they have been mostly associated with farming and industrial operations. They usually exert a very high oxygen demand and they can cause failure of on-site wastewater treatment components sized solely on HLRs.

5.1 Materials and Methods

5.1.1 Alum Sludge-Based CW System

Figure 5.1 shows the schematic diagram of the laboratory-scale CW. The system consists of four stages of individual CWs, which are all linked together using peristaltic pumps. Dewatered alum sludge was collected and characterised as described in Section 3.1.1. The collected alum sludge was air dried and filled into each stage of the CW system up to a depth of 50 cm. The air-dried alum sludge used had a d_{10} and d_{60} of 0.5 mm and 1.8 mm respectively. Each stage had 10 cm of 6 to 10 mm gravel at the base to serve as support. Young *Phragmites australis* obtained from a local supplier was planted on top of each stage.

5.1.2 System Operation, Sampling and Analytical Methods

The CW was operated using the tidal flow strategy. 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 consisted of 1 hour of wastewater contact with the alum sludge and 3 hours of resting, during which the wastewater was drained out from the CW and the system allowed to rest before the next cycle commenced. Wastewater collected from an animal research farm was used as influent into the CW. Appropriate dilution of the wastewater was carried out as necessary. The wastewater was then loaded sequentially in batches onto the system, from the first stage to the last stage, using peristaltic pumps at an HLR of 1.27 m^3/m^2.d. Samples were collected periodically from the feed tank (influent) and from the outlet of each stage of the system (effluents). These samples were promptly analysed for BOD_5, COD, SS, turbidity, PO_4-P (both RP and SRP), pH, NH_4-N, NO_2-N, NO_3-N and total nitrogen (TN) according to standard methods. In order to capture the behaviour of the CW during the monitoring stage, a neural network-based pattern analysis technique known as self-organising maps (SOM) was used to mine data on BOD_5, COD, RP and SRP. Comprehensive details of the application of the SOM technique can be found in Hong et al. (2003), Aguado et al. (2008) and Akratos et al. (2009).

The SOM was applied to detect and visualise the pattern and relationship between the removal of organics and P during the monitoring period. Furthermore, in order to estimate the consumption of DO for microbial oxidation in the system, and therefore provide a means of examining the tidal flow concept, the DO profile was monitored in the headspace of Stages 1 and 2 of the CW. The profiling was conducted before, during and after feeding the system with wastewater.

5.1.3 Calculation of the Theoretical Oxygen Input by the Tidal Flow Strategy

A tidal flow strategy enhances oxygen input into the CW based on convective transport, and the ultimate sizing and design of the tidal flow system will be intimately linked to the oxygen-transfer capacity of the system. The theoretical oxygen transfer based on the tidal flow strategy used in this study was determined as follows:

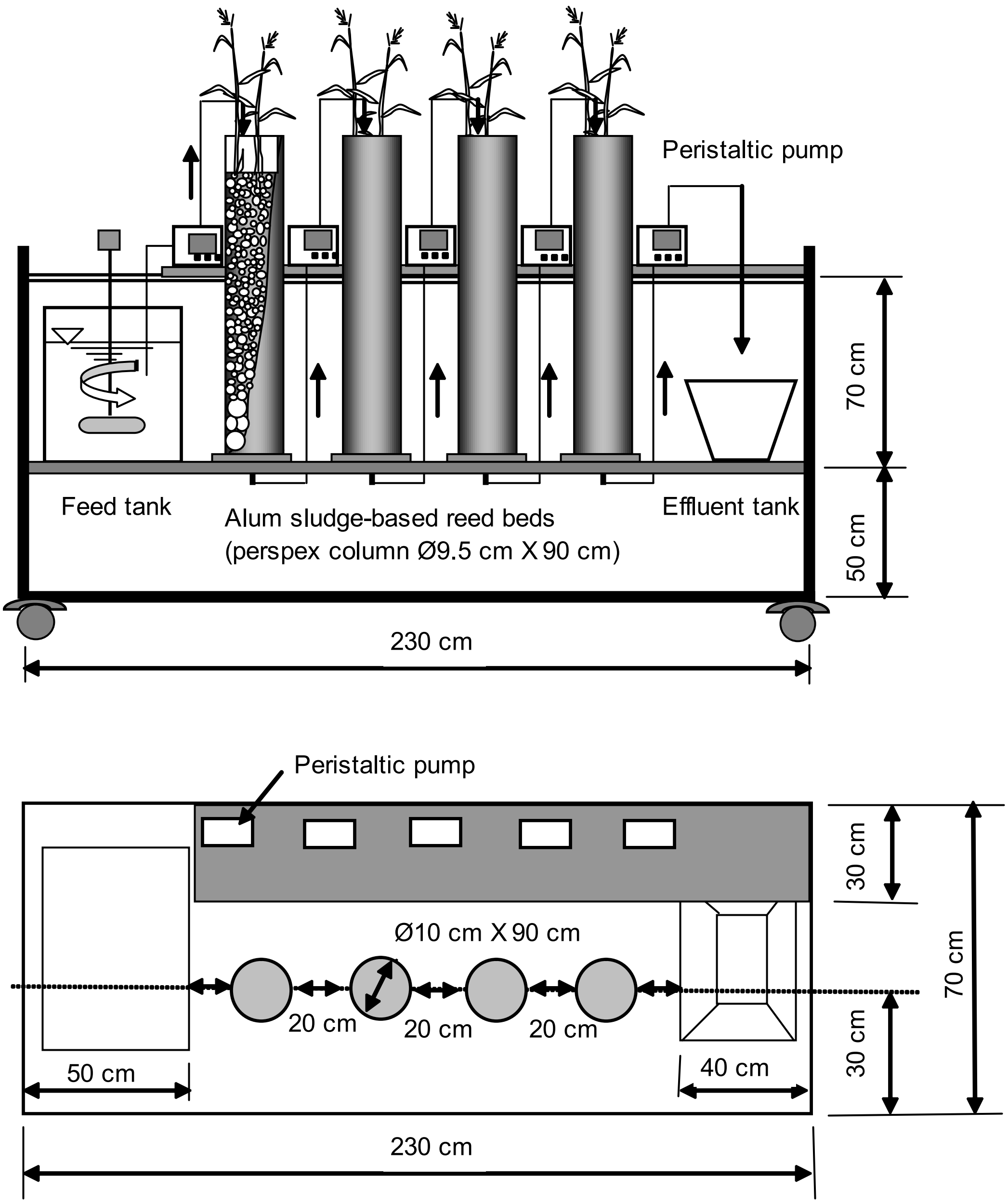


Figure 5.1. Layout of the 4-stage alum sludge-based constructed wetland system.

A pore volume of 1.5 L was used in the design to ensure subsurface flow, and this is ca. 75% of the pore volume in each cell of the CW. Therefore, the mass of the air drawn in in each cycle = 1.5 L × 1 .2 g/L = 1.8 g, and the mass of O_2 drawn in = 21% × 1.8 g-O_2 = 0.378 g-O_2.

Thus, a value of 0.378 g-O_2 is obtained and this represents the theoretical amount of oxygen that is drawn into each individual stage of the treatment system in each tidal cycle. This value can be multiplied by the number of stages in the CW to determine the total amount of oxygen drawn into the system. By having an estimate of the percentage of DO available, the amount of organic load that can be removed in the entire system can be reasonably determined from Table 5.1. In addition, by determining the amount of organics removed in the system, the oxygen-transfer efficiency of the system can also be calculated.

Table 5.1. Estimation of system's treatment capacity.

[a]% DO available	[b]Equivalent DO available (in mg of O_2)	[c]Oxidisable BOD_5 (mg/L)	[d]Filterable BOD_5 (mg/L)	[e]Stage removal capacity (mg/L)	[f]System removal capacity (mg/L)	O_2 transfer capacity (g-O_2/m^2.cycle)
100	378	378	378	756	3024	53.3
75	283.5	283.5	283.5	567	2268	39.9
50	189	189	189	378	1512	26.7
25	94.5	94.5	94.5	189	756	13.3
10	37.8	37.8	37.8	75.6	302.4	5.3
5	18.9	18.9	18.9	37.8	151.2	2.7
1	3.78	3.78	3.78	7.56	30.24	0.5

[a] refers to the potential percentage of the available oxygen that is converted to dissolved oxygen (DO) for microbial oxidation, [b] refers to the equivalent DO available, calculation based on the % of DO available and a pore volume of 1.5 L, [c] refers to the amount of organics removable from the wastewater by microbial oxidation, [d] refers to the amount of organics removable from the wastewater by filtration, [e] refers to the total amount of organic load removable from the wastewater in each stage per tidal cycle, [f] refers to the total amount of organic load removable from the wastewater in the entire system in each tidal cycle, based on a 4-stage system. All values are based on a single fill and drain cycle operation.

5.2 Results and Discussion

5.2.1 *General Treatment Performance*

Figure 5.2 shows the removal trend for organics (BOD_5 and COD) and P (RP and SRP) across the entire system over the monitoring period. For the organics, the system achieved higher removal efficiencies for BOD_5 than for COD. However, this is quite possible as it can be noted from Fig. 5.2 that the ratio of the BOD_5 concentration to the COD concentration (C_{BOD}/C_{COD}) is high and this represents an effluent that should be highly amenable to a biologically based treatment. With mean influent values of 392.7 ± 95.6 mg/L for BOD_5 and 579.8 ± 142.0 mg/L for COD, average removal efficiencies of 90.6 ± 7.5% and 71.8 ± 10.2% for BOD_5 and COD, respectively, were achieved. However, the average C_{BOD}/C_{COD} in the effluent exiting the system was 0.23, indicating that a high level of treatment has been achieved. The system achieved a comparable performance with other similar systems: for example, Sun et al. (1999) reported mean removal efficiencies of 97.6% and 71.3% for BOD_5 and COD respectively, for a combined tidal-flow down-flow system with multiple sub-beds. The reported sub-beds had areas ranging from 2.56 m^2 to 3.25 m^2 each and this would result in a lower overall loading (as compared to the loading used in this study).

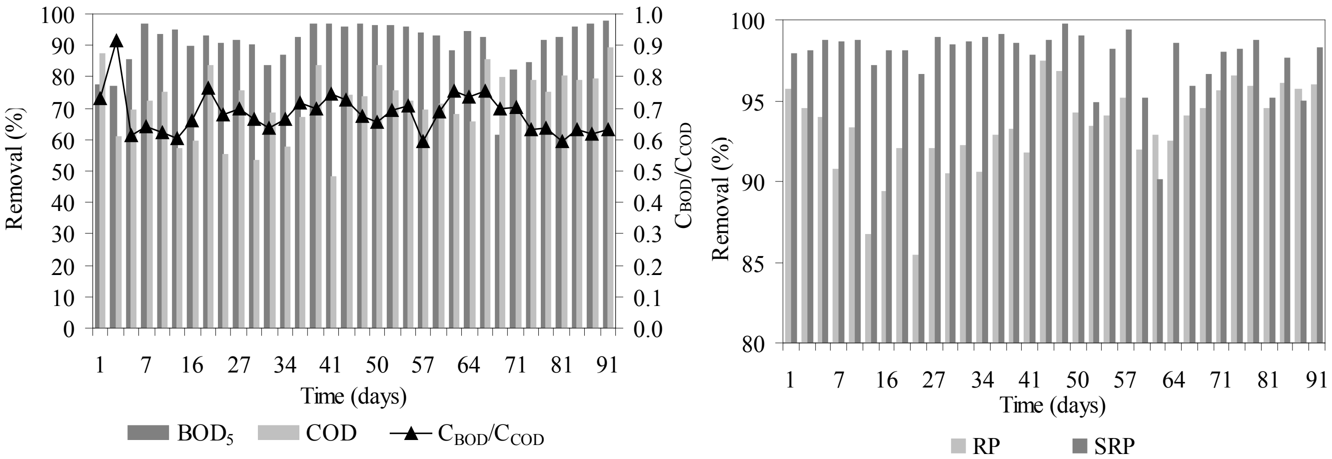


Figure 5.2. Trend of pollutant removal efficiencies in the novel constructed wetland system RP = reactive phosphorous, SRP = soluble reactive phosphorous.

Cerezo et al. (2001) also reported removal efficiencies of 90% and 70% for BOD_5 and COD respectively for a field-based CW made of eight tanks in three series and with a surface area of 1 m^2. In another study, Zhao et al. (2004) used an HLR of 0.43 $m^3/m^2.d$ (≈ one-third of the HLR used in this study) in a gravel-based tidal-flow CW, and achieved an overall treatment efficiency of 77.3% (COD) and 77.7% (BOD_5). While the system in this study had a relatively smaller footprint and higher loading, it can be seen that the performance obtained in the present treatment system is comparable and promising.

Phosphorus removal in the system was exceptional, and in particular the system proved very effective for the removal of SRP. In a CW, it is often a challenge to achieve concurrently high removal efficiencies for P and organic matter (BOD_5, COD). Often it is found that the efficiency of P removal is low compared to other parameters such as BOD_5 (Park, 2009). In the current system, with mean influent P values of 21.0 and 45.3 mg-P/L for SRP and RP respectively, removal efficiencies of 97.6 ± 1.9% (SRP) and 93.3 ± 2.7% (RP) were achieved indicating a distinctive and superior P removal performance, especially when compared to P removal in CWs reported in the literature. The results of the SOM mapping for data on BOD_5, COD, SRP and RP are shown in Fig. 5.3. From this figure, it can be determined that the lower part of the BOD-in map represents the high BOD-in values as depicted from the BOD-in component plane. This shows that the states of the high influent BOD values are mapped into that part of the map. However, through visual investigation of the BOD-out component plane, the BOD-in component is mainly negatively correlated with the BOD-out component.

For further illustration, it can be seen that the lower part of the BOD-in map (as depicted from the BOD-in component plane) is mainly matched with low influent BOD_5 values as can be seen from the colour code of the lower part of the BOD-out map and the represented value read from the component plane. In essence, the BOD mapping suggests that even with higher influent BOD values, the effluent BOD values are still low. For the COD mapping, it can be observed that the lower part of the COD-in map has some slight positive correlation with high COD-out values as depicted in the colour code of the lower part of the COD-out map. As has been discussed in Section 5.2.1, the COD values in the final effluent from the system were relatively higher

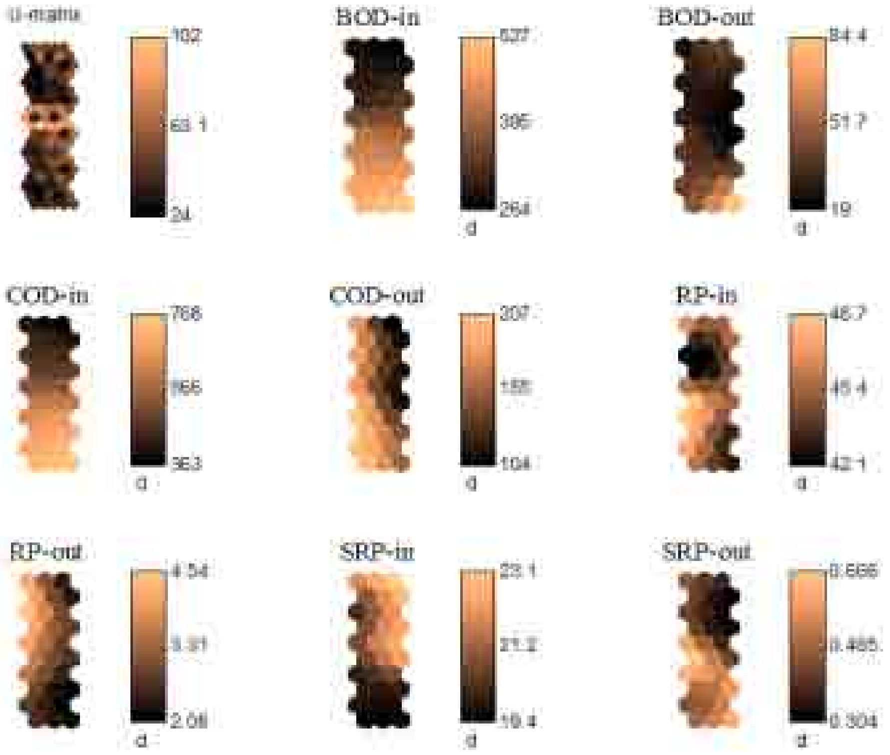


Figure 5.3. Abstract visualisation of the relationship between influent and effluent values of BOD_5, COD, reactive phosphorus (RP) and soluble reactive phosphorus (SRP) in the constructed wetland system using self-organising maps.

than the BOD_5 values. However, the average BOD_5/COD value of the final effluent was 0.23, indicating that most of the organics in the wastewater have been degraded in the CW, leaving a final effluent with a very low biodegradability.

However, for both RP and SRP, it is interesting to observe that the local average values for both RP-out and SRP-out were very low, irrespective of the influent levels. The exceptional ability of the system to achieve low effluent P concentrations irrespective of the influent P concentration can be attributed to the adsorption ability of the alum sludge used as the substrate. There was a slight reduction in the overall removal of TN, with an average overall removal of 22.8% obtained. However, NH_4-N was reasonably well removed in the system, with an average overall removal of 73.2% obtained.

The SS concentration in the influent wastewater ranged from 88 to 553 mg/L. However, the system removed significant amounts of solids from the influent wastewater with a removal efficiency (mean ± SD) of 89.3 ± 7.7% being achieved. Final effluent SS concentrations ranged from 3 to 45 mg/L. Similarly, the turbidity levels were also reduced significantly with a removal efficiency of 86.7 ± 9.2% achieved.

5.2.2 Inter-Stage Treatment Performance

In order to examine the role of individual stages in pollutants removal, the removal fraction in each stage was determined as a percentage of the overall removal achieved in the system, and shown in Table 5.2 alongside the respective concentration in each stage. The most important feature from Table 5.2 is the high removal fractions obtained in the first stage, irrespective of the pollutant. This reflects the key role of the first stage in the overall removal. For the organics, the highest reduction of 53% (BOD_5) and 39% (COD) in both cases occurred in the first stage, while the reduction in the last stage (Stage 4) was the least. The individual contribution of the subsequent stages ranged from 6 to 23% (BOD_5) and 7 to 16% (COD). The high efficiency of the first stage may be explained by the rapid degradation of easily degradable pollutants in the raw wastewater that flows through and interacts with the bed matrices and biofilms (Tchobanoglous et al., 2003). In terms of P removal, there was no significant P removal beyond the first stage for both SRP and RP, as can be seen in Table 5.2. The rapid removal of P from the influent wastewater, particularly in the first stage, is attributable to the ability of the alum sludge to remove P. Both SRP and RP were reduced substantially in the first stage, leading to insignificant reductions in the subsequent stages. It is believed that the P removal will be significantly performed in the subsequent stages when the first stage becomes saturated. The removal of SS and the reduction of turbidity followed the same trend, with the highest reduction obtained in the first stage. Although there were further reductions in the subsequent stages, their relative contributions were less significant.

Table 5.2. Overall and inter-stage treatment performance in the system.

Parameter	Influent	Stage 1	Stage 2	Stage 3	Stage 4	% Removal
BOD_5	392.7	184.1 (53)	93.7 (23)	60.4 (9)	37.1 (6)	90.6
COD	579.8	353.7 (39)	260.7 (16)	200.1 (11)	158.1 (7)	71.8
SRP	21	2.3 (89)	1.2 (5)	0.86 (2)	0.47 (2)	97.6
RP	45.3	8.9 (80)	5.7 (7)	3.7 (4)	3.0 (2)	93.3
SS	218	72.9 (67)	39.8 (15)	29.2 (5)	19.3 (5)	89.3
Turbidity	127.7	46.4 (64)	26.5 (16)	18.6 (6)	14.3 (3)	86.7
TN	142.5	133.8 (64)	125.6 (6)	120 (4)	110 (7)	22.8
NH_4-N	132.9	87.1 (35)	46.8 (30)	40.2 (5)	35.6 (4)	73.2
NO_2-N	0.75	0.23	0.13	0.06	0.15	–
NO_3-N	3.6	9	27.2	9.5	25	–
pH	7.81	7.0	6.82	6.82	6.81	–

The profile of nitrogen (N) removal in the system can also be seen in Table 5.2. As previously discussed, NH_4-N was reasonably removed in the system with an average overall removal of 73.2% obtained. However, there was an increase observed in the level of NO_3-N as the wastewater passed through the system. The highest increase in NO_3-N values was in Stage 2 when the level increased from 9 mg/L in Stage 1 to 27.2 mg/L in Stage 2. The NO_3-N levels in the subsequent stages remained higher than the level in the influent, but they were lower than the level obtained in Stage 2. This suggests nitrification was occurring as the nitrifying bacteria converted NH_4-N in the influent to NO_2-N and further to NO_3-N. However, while there was evidence of nitrification in the system, as suggested by the increase in NO_3-N levels across the stages, a corresponding decrease in pH, as would be expected, was not observed. As can be seen from Table 5.2, the pH decreased from 7.8 in the influent to 7.0 at the exit of Stage 1.

All parameter values are in mg/L, except pH (no unit) and turbidity which is in nephelometic turbidity units (NTU). All P values are in mg-P/L. Bold values in brackets refer to the amount of removal in percentage obtained in each respective stage, relative to the initial concentration in the wastewater and expressed as a percentage.

However, although the pH decreased further to 6.82 after exiting Stage 2, there was no further pH decrease in the subsequent stages. A possible explanation for this is that, while indeed there may be a pH decrease across the stages as a result of nitrification, such a decrease may be masked by the buffering effect of the alum sludge used.

5.2.3 *Dissolved Oxygen Profiling*

The profiling of oxygen in the headspace of the system is useful for examining the mechanism and capacity of oxygen transfer by the tidal flow strategy. The profiling is divided into four phases. Phase ‘a’ refers to the period just before the pumps are operated to begin the filling process. In this phase, the system headspace is in equilibrium with the atmospheric air. However, before the pump-in begins, the system is made airtight, and the profiling begins.

Phase ‘b’ refers to the period during which the wastewater is in contact with the alum sludge bed in the system. Phase ‘c’ refers to the short period during which the wastewater is being drained out, and Phase ‘d’ refers to the period after which the bed has been drained out and the system is being rested. The profiling was carried out for Stages 1 and 2 in the treatment system after 6 weeks of operation. Results show that the percentage decrease in oxygen was higher in Stage 1 than in Stage 2. This means that, although, based on geometrical calculations, a potentially equal volume of air is drawn into the head space of both stages, more DO is used in the first stage than in the second. This suggests more biomass development in the first stage than in the second stage. Consequently, the amount of DO used is higher in Stage 1 than in Stage 2.

Furthermore, based on the trend in the percentage of oxygen concentration in Stages 1 and 2, the following suggestions can be made. Prior to the pumping of the wastewater, the headspaces in the respective stages were equilibrated with the atmosphere, and consequently the headspace was saturated with air, which is composed of 21% oxygen. This is depicted as Phase ‘a’. During and after the pumping (i.e. Phase ‘b’, which is also the period of contact of the system with wastewater), there was a minor drop in the oxygen concentration in the system. The drop was quite small and it could have been caused by air drift, instrument error, or it is possible that actual microbial-degradation was taking place. Phase ‘c’ is the pump out and any change noticed during this period was probably caused by the suction effect of the draining wastewater. However, the most significant drop in DO concentration was during Phase ‘d’, when the respective stages were at rest. This suggests the utilisation of oxygen by the microorganism for bacterial oxidation.

Overall, the decrease in oxygen concentration was ca. 16% and 10% in the first and second stages respectively. By calculation, a 16% change in the oxygen concentration would imply that 40.3 mg/L of oxygen was utilised by the microorganisms in the first stage. If this is the case, then, up to 80.64 mg/L of BOD_5 can be removed in the first stage, and, based on this, a reasonable estimate can be made for the subsequent stages and the entire system, assuming the same amount of oxygen is utilised in each stage after biomass establishment. Using this result, it would appear that almost the entire organic load introduced into the system is removed, and, by implication, 10 to

25% of the oxygen input is available as DO for microbial oxidation (see Table 5.1 above). This puts the oxygen-transfer efficiency of the current system under study at between 5.3 and 13.3 g-O_2/m^2. cycle.

5.3 Summary

A novel multistage CW using alum sludge as the main substrate was conceptualised, designed and operated to enhance the concurrent removal of P and organic matter (OM) from wastewater. The system achieved very high removal efficiencies for BOD_5 (90.6%), COD (71.8%), RP (80%) and SRP (89%). In all cases, the first stage of the CW contributed most significantly to the overall removal of the pollutants, particularly in the case of SRP where 89% of the overall removal was achieved in the first stage compared to 53%, 39% and 80% removal achieved for BOD_5, COD and RP respectively in the first stage. Interestingly, the CW was able to achieve concurrently high removal efficiencies for P and OM because of the combination of the alum sludge as main substrate and the use of the tidal flow strategy. There was apparent nitrification in the system, but no marked decrease in pH was observed.

6 Pilot-Scale Trial: Field Demonstration Study

After extensive laboratory-scale investigations, a pilot-scale field study was conducted on an animal farm with real wastewater for 10 months. The purpose of the field study was to: (i) validate the achievement obtained in the laboratory study, and (ii) gain experience from the engineering point of view especially for the possible large-scale application of the CW.

6.1 Materials and Methods

The field-scale system was constructed on a farm in Newcastle, Co. Dublin, Ireland. It consisted of four treatment stages (see Fig. 6.1) constructed using similar 1100 L plastic bins. The stages were all linked together to form the system using pipes which were also connected to a 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 samples. For the configuration, each stage of the system had 10 cm of 10 mm gravel at the bottom to serve as support layer followed by 65 cm of the alum sludge cakes as main substrate layer and then 10 cm of 20 mm 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 the Ballymore-Eustace Water Treatment Plant in south-west Dublin, Ireland where aluminium sulphate is used as coagulant (also see Section 3.1.1). The size/length (mean±SD) of the alum sludge cakes used was 3.19±0.89 cm.

Common reed, *Phragmites australis*, was planted on top of each stage. The system was operated as a subsurface flow engineered wetland system and a tidal flow operation strategy was used. Owing to the variable nature of wastewater generated on the farm and the need to operate the system with gradually increasing wastewater strength to allow for the system to stabilise and the reeds to grow, wastewater from the farm activities was first collected from the holding tank on the farm and pumped into a 10,000 L capacity tank. Appropriate dilution was then carried out to achieve the desired concentration. Thereafter, the prepared wastewater was gravity fed into an underground tank (with a ball-float valve control) which served as the influent tank from where the wastewater was pumped into the alum sludge-based CW system. There were three cycles per day and each cycle included 4 hours of wastewater contact in each stage and 4 hours of rest, during which wastewater was drained out (to the next stage) and the stage 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 periodically and analysed for COD, BOD_5 TP, $PO_{4\text{-}}P$, TN, $NH_{4\text{-}}N$, $NO_{3\text{-}}N$, $NO_{2\text{-}}N$, SS, turbidity, aluminium (Al) and soluble Al according to standard methods.

Figure 6.1. Layout of the pilot-scale field alum sludge-based constructed wetland system.

6.2 Results and Discussion

6.2.1 Organic Matter

The overall COD removal performance of the system is illustrated in Fig. 6.2. During the start-up period (25/02/2009 to 02/06/2009), the influent COD ranged from 57 to 1087 mg/L with an average of 463 mg/L, while the effluent COD ranged from 97 to 790 mg/L. During this period, the COD removal was low as biological activity was being developed. The second period began on 5/6/2009 with pig slurry being used as the influent. The influent COD ranged from 310 to 1578 mg/L during this period, while the effluent COD averaged 270 mg/L. With pig slurry, the COD removal performance was improved significantly with average removal efficiency of 67.7%. It seems that the effect of temperature on COD removal is very limited. The removal efficiency remained around 67% even when the temperature dropped below 6°C in December 2009 as shown in Fig. 6.2. The average BOD removal efficiencies of 51.4% and 66.8% were recorded for the start-up and the second period, respectively, as

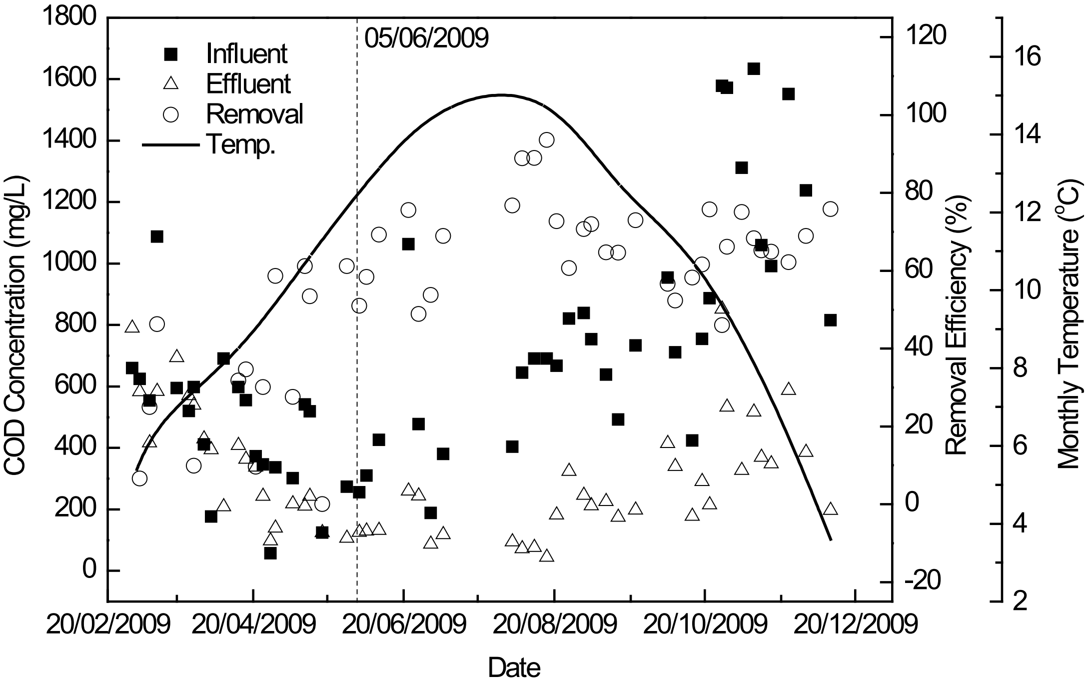


Figure 6.2. COD removal and monthly temperature.

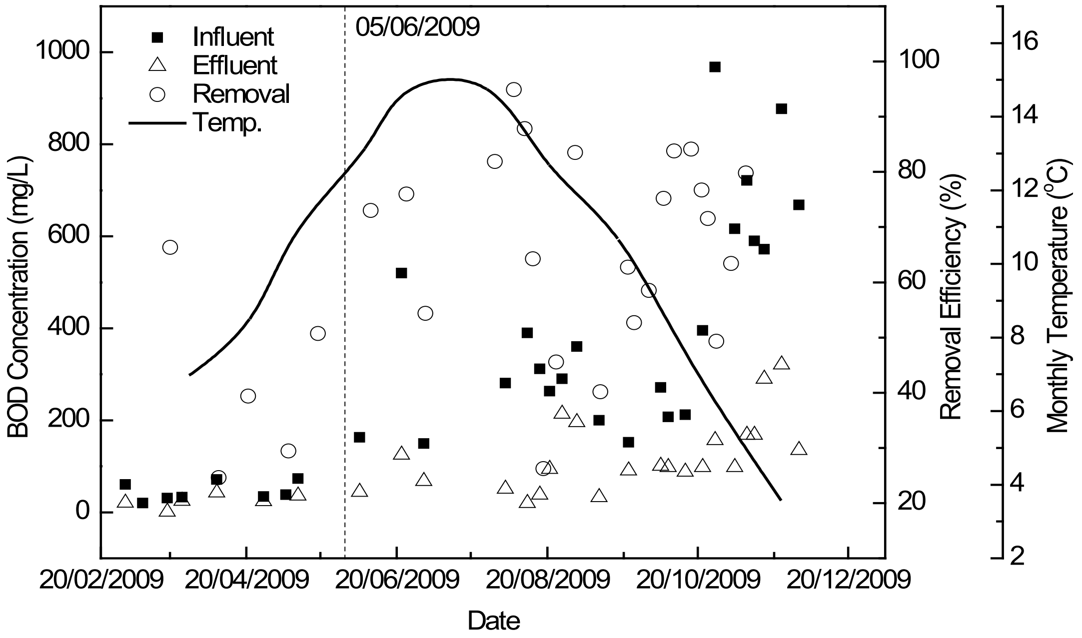


Figure 6.3. BOD removal and monthly temperature.

shown in Fig. 6.3. It is noted that the BOD removal showed some scatter, probably due to the monitoring facility.

6.2.2 *Nitrification Performance*

The overall nitrification performance is shown in Fig. 6.4. Nitrification became established after about 2 months. Ammonia removal efficiency of 87% was obtained on 24 April. Between 24 April and 9 November, very stable and efficient nitrification was achieved with an average ammonia removal of 89.2%. After 12 November, the nitrification decreased sharply from over 90% to 48%.

This can be explained by the fall of the temperature to <6°C, because the nitrification autotroph has a low specific grow rate and therefore its growth is very sensitive to temperature.

6.2.3 *Total Nitrogen Elimination*

Figure 6.5 illustrates the overall TN removal performance. The TN elimination is largely dependent on the influent BOD/TN ratio, i.e. the available carbon source, as demonstrated in Fig. 6.5. During the start-up period, the influent BOD/TN ratio was very low with an average of 0.9. According to Henze et al. (1997), for complete denitrification the influent BOD/TN ratio should be close to 5. Apparently, the influent BOD/TN ratio during the start-up period was far lower than this value. Consequently, an average TN removal efficiency of only 27% was achieved during this period. From 5 June to 17 August, the influent BOD/TN ratio increased significantly with an average of 2.41 because of the nature of the influent. As a result, the TN removal increased significantly from 50% to over 90%, with an average of 64% removal over this period. However, from 21 August to 22 October, the influent BOD/COD again decreased and this affected the TN removal, which dropped to 13.5 to 47.8%. The rapid response of TN removal efficiency to the increase of the influent TN/COD ratio was observed again during the period between 27 October and 23 November. However, the drop after 23 November cannot be explained as being caused by an insufficient carbon source, because the BOD/TN ratio was still above 4. Instead, it tracked the decrease in nitrification performance (Fig. 6.4), that is, the TN elimination was limited by the nitrification performance which in turn was caused by the drop in temperature. All these results indicate that high TN elimination can be achieved within the CW, provided that the carbon source is sufficient for denitrification.

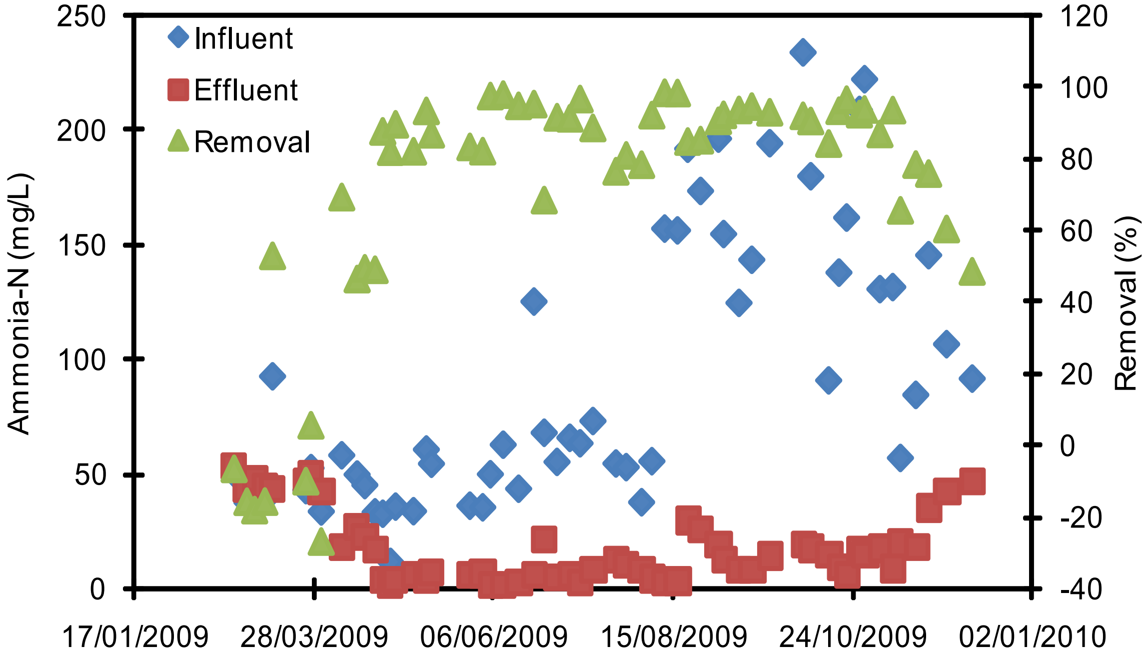


Figure 6.4. Nitrification performance and monthly temperature.

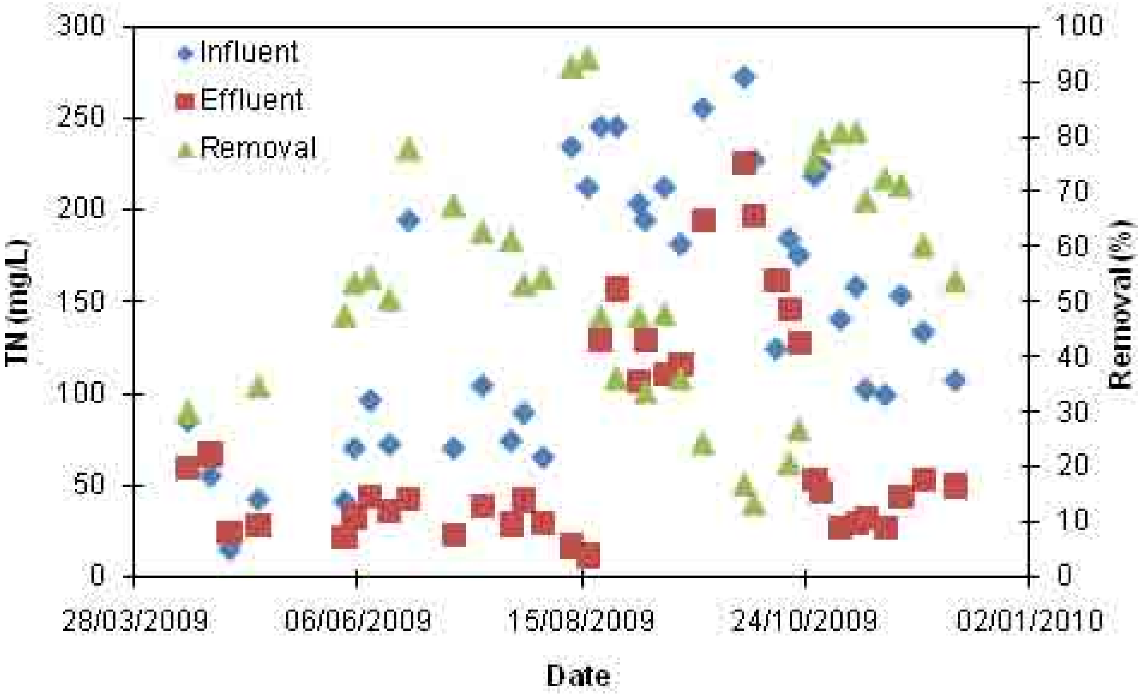


Figure 6.5. Total nitrogen (TN) nitrogen removal and influent BOD/TN ratio.

6.2.4 Phosphorus Removal

Performance of the CW system demonstrates a high and stable P removal, as illustrated in Fig. 6.6. A removal efficiency of above 90% was achieved within 1 week of operation. Thereafter, the removal efficiency was maintained in the range of 82 to 100% under a variable P loading rate caused by the nature of the influent wastewater. Removal efficiencies remained above 90% most of the time, with the average being 94.6% up to 12 November. The main pathway of P elimination is believed to be adsorption on the main substrate (i.e. alum sludge). However, from 12 November the removal efficiency started to decrease and dropped to 73% on 10 December 2009. This may be because the alum sludge was starting to get saturated, since the P adsorption on the alum sludge has continued for nearly one 1 year under a high P loading rate. This assumption can be confirmed during further operation of the CW.

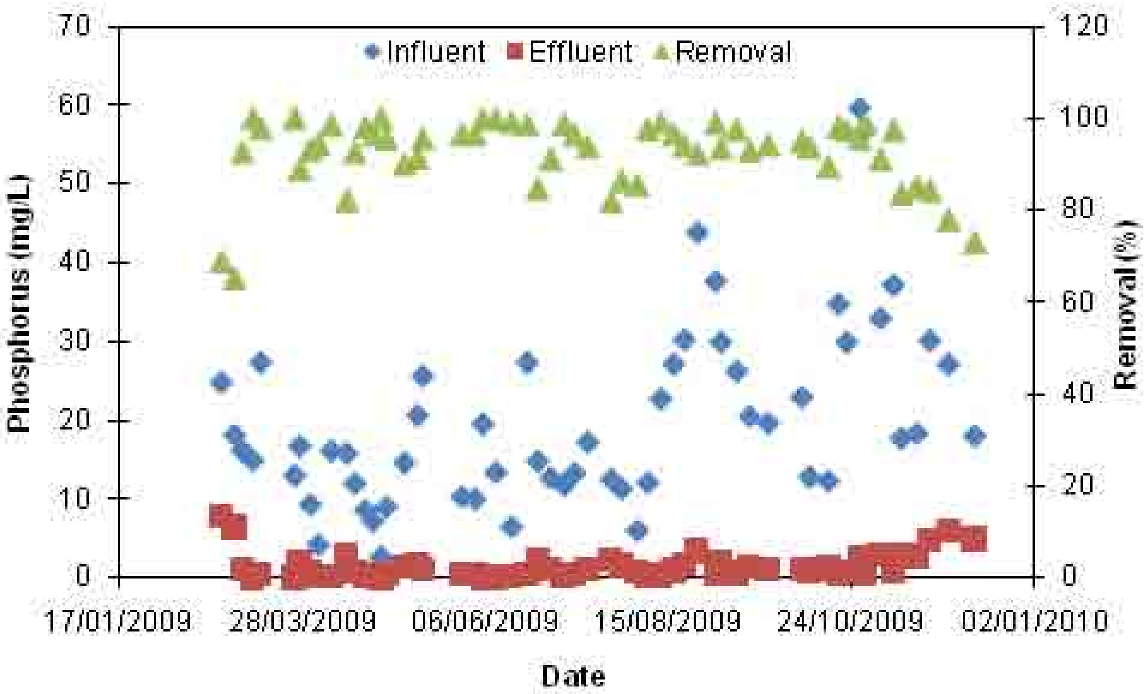


Figure 6.6. Phosphorus removal and monthly temperature.

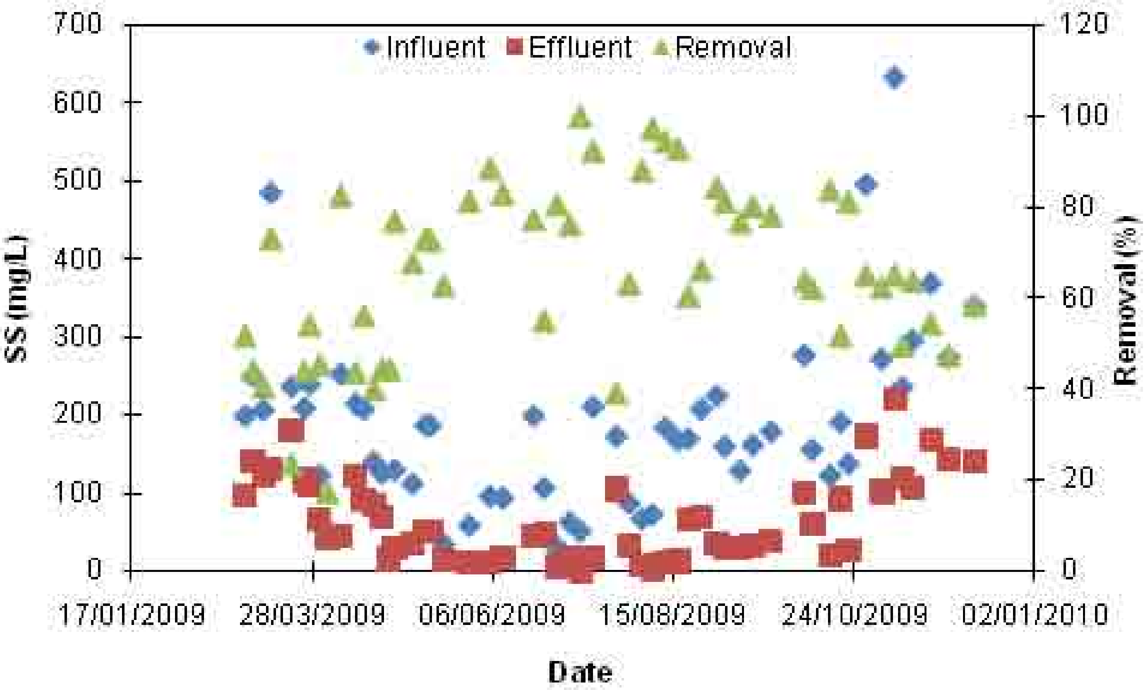


Figure 6.7. Suspended solid (SS) removal.

6.2.5 *Suspended Solid Removal*

Figure 6.7 illustrates the overall SS removal performance. During the start-up period, the influent SS ranged between 27 and 485 mg/L, with an average of 175 mg/L. The effluent SS was between 11 and 181 mg/L, with an average of 77 mg/L. Accordingly, the removal efficiency varied between 17 and 83%, with an average of 56%. In the second operation period with pig slurry from 5 June, the influent and effluent SS ranged, respectively, between 31 and 633 mg/L with an average of 195 mg/L, and between 0 and 221 mg/L, with an average of 62 mg/L.

6.2.6 *Inter-Stage Performance*

Figure 6.8 depicts a typical inter-stage performance of a cycle in the tidal flow (on 22 June 2009) of organic matter, N and P removal. The figure shows that COD and soluble COD removal mainly occured in Stage 1 to Stage 3 (Fig. 6.8a); 26%, 23% and 22% of influent COD were removed in Stages 1, 2 and 3, respectively, while Stage 4 accounted for only 9% of the COD reduction. Regarding BOD removal, Stage 2 accounted for 51% of the BOD reduction (Fig. 6.8a), and the pattern of TN and NH_4^+-N elimination was similar to COD removal (Fig. 6.8b). It should be noted that there was no nitrate accumulation in Stages 1 and 2, that is, all the oxidised ammonia was eliminated through denitrification. Further TN reduction was also observed in Stage 3. This suggests that simultaneous nitrification and denitrification (SND) can be the main pathway of TN removal because of the oxygen diffusion limitation within the biofilm. However, no further N reduction was observed in Stage 4 owing to a lack of a carbon source. To stimulate SND and therefore further improve the TN removal performance, step feed configuration can be considered. Multiple anoxic/aerobic stages with or without recirculation configurations are also possible options. Unlike organic matter and N removal, P elimination was observed mainly in Stage 1, which accounted for 67% of P reduction (Fig. 6.8c).

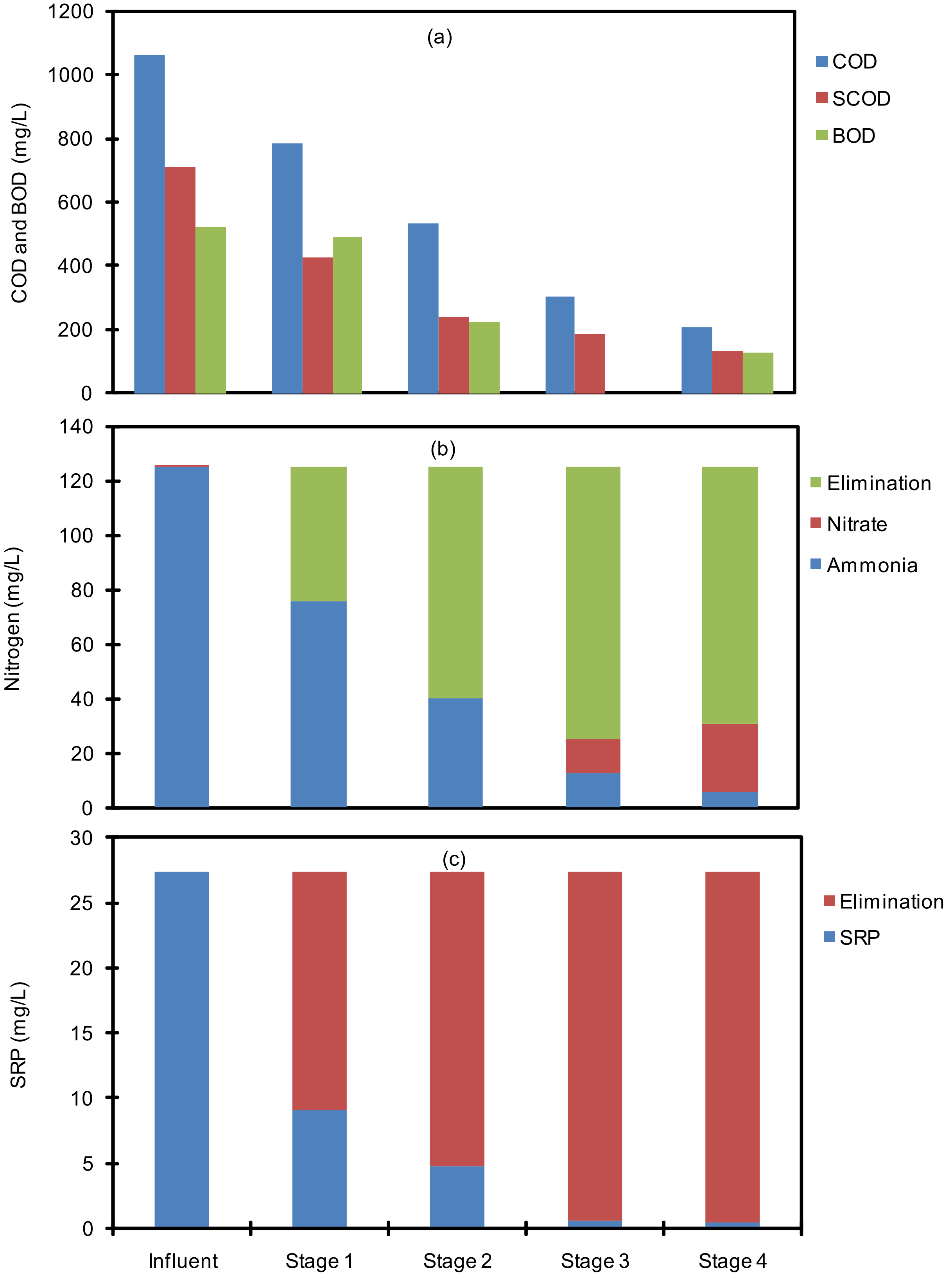


Figure 6.8. Cycle performance in (a) organic matter (COD and BOD), (b) nitrogen and (c) soluble reactive phosphorus (SRP), 22 June 2009, sCOD = soluble chemical oxygen demand.

6.3 Summary

The first pilot-scale field system of alum sludge-based CW has demonstrated that the system is a unique and promising low-cost wastewater treatment. Although technical parameters have been obtained from the field trial, which are useful for the purpose of design and operation, a longer-term operation is still desirable to validate the stability of the system. In particular, the clogging of the system during the long-term operation should be tested and the saturation of the alum sludge with P might also be usefully investigated over a more extended period before the large-scale application of the system.

7 Modelling the Novel Constructed Wetland Treatment Behaviour

The modelling of wastewater treatment processes has long been of interest to environmental engineers, and the modelling of such processes in CWs is no exception. However, given the multiplicity and variable nature of reactions occurring in CWs, the quantification of all specific rate-controlling mechanisms may be rather difficult. Nonetheless, several mathematical models have been proposed (Mayo and Bigambo, 2005; Nabizadeh and Mesdaghinia, 2006; Langergrager et al., 2009; Pimpan and Jindal, 2009), but they are mainly for horizontal flow CWs. Given that a tidal flow CW is a relatively new concept in CW development, there is not yet any significant research towards mathematical modelling of its treatment performance. A specific mathematical modelling approach was adopted in this study in an attempt to enable the prediction of the treatment performance and assist in establishing a performance model. However, the model can be described as producing only a moderate fit between expected and predicted values.

7.1 Mathematical Modelling

As a preliminary attempt, a mathematical modelling approach previously developed by Sun et al. (1998) was adopted in this study.

The equilibrium in the adsorption process can be defined as:

$$\text{surface}(H_2O)_x - \text{adsorbate}(H_2O) \leftrightarrow \text{surface} - \text{adsorbate}(H_2O)_z$$

The surface refers to the adsorbent surface. The relationship between the components in this equilibrium can be expressed in the form of several adsorption isotherms. However, the Freundlich isotherm equation (Eq. 7.1) is employed here (Sun et al., 1998):

$$\frac{x_o}{m} = K_o C^{1/n} \tag{7.1}$$

Where x_o is the amount of pollutant adsorbed (mg), m is the amount of adsorbent (mg), K_o is the equilibrium constant indicative of the strength of adsorption, C is the equilibrium concentration of the pollutant in solution (mg/L) and $1/n$ is the degree of non-linearity. Equation 7.1 can further be expressed as Eq. 7.2 to represent all the pollutants in the wastewater instead of a single specified pollutant:

$$\frac{x}{m} = K_1' C'^{1/n} \tag{7.2}$$

Where x is the amount of the adsorbed pollutants, expressed as the oxygen demand for their biological decomposition (mg), K_1' is the equilibrium constant indicative of the strength of adsorption, and C' is the equilibrium BOD_5 value of the wastewater (mg/L). Generally, the value of $1/n$ often approximates to 1 (Sun et al., 1998), and consequently Eq. 7.2 can be expressed in Eq. 7.3 as:

$$x = mK_1'C = A_h h \rho K_1' C = A_h h K_1 C' \tag{7.3}$$

Where A_h is the wetland area (m^2), h the wetland depth (m), ρ the superficial density of the wetland (mg/m^3) and $K_1 = \rho K_1'$. The pollutant removal from wastewater by adsorption during the feeding process can be described by Eq. 7.4:

$$C_1 = C_o - \frac{x - x'}{v_f} \tag{7.4}$$

Where C_O is the influent BOD_5 value (mg/L), C_1 the effluent BOD_5 value (mg/L), x' the oxygen demand of the pollutants adsorbed in the wetland matrix before the feeding pulse (mg) and v_f the volume of wastewater during each feed (L). During the rest stage, atmospheric oxygen diffuses into the bed matrix, and the retained pollutants are aerobically decomposed by microorganism in a process usually regarded as a first-order reaction process. Thus, the reaction rate can be expressed in Eq. 7.5 as:

$$-\frac{dC_{film}}{dt} = K_2 C_{film} \tag{7.5}$$

Where C_{film} is the BOD_5 content of the biofilm on the wetland matrix (mg/L), t is the time (h) and K_2 the reaction constant (h^{-1}).

By defining $C_{film} = x / V_{film}$, where V_{film} is the volume of the biofilm on the wetland matrix (L) and is assumed to be constant (Sun et al., 1998), then Eq. 7.6 is:

$$\frac{dx}{dt} = K_2 dt \tag{7.6}$$

By taking integrals and solving with respect to the boundary limits, as in Eqs 7.7 and 7.8:

$$\int_x^{x'} -\frac{1}{x}dx = \int_o^{t_o} K_2 dt \tag{7.7}$$

$$x' = \exp\left(-K_2 t_o\right) \tag{7.8}$$

Where to is the time interval between two cycles (hours [h]). By substituting for x' and x (from Eq. 7.8 and Eq. 7.3 respectively) and applying these substitutions in Eq. 7.4,

$C_1 = C_o - \dfrac{A_h h K_1 C_1 - A_h h K_1 C_1 \exp(-K_2 t_o)}{v_f}$ and this gives Eq. 7.9:

$$C_1 = \frac{C_O v_f}{v_f + A_h h K_1\left(1 - \exp\left(-K_2 t_o\right)\right)} \tag{7.9}$$

Equation 7.9 was solved using Microsoft Excel Solver package based on the dataset collected during the first phase of operation of the current system. The solved equation gives the optimal values of the constants K_1 (which indicates the adsorption factor of the wetland) and K_2 (which indicates the reaction constant and which is specific for the effluent being treated). The optimal values of these constants represent the optimal values of the constants that can be used in the mathematical model to predict the effluent concentration of the current system under study. In the future, such a model, when validated, can be used for design purposes. A comparison was made between the predicted and observed (experimental) effluent concentration values and the percentage deviation (PD) was also calculated using Eq. 7.10:

$$\mathrm{PD} = \frac{C^{\text{predicted}} - C^{\text{observed}}}{C^{\text{observed}}} \times 100 \tag{7.10}$$

7.2 Experimental Set-up and Model Tests Results

The experimental set-up and operation have been described in Section 5.1.1 and the performance of the CW system has been presented in Section 5.2. Based on data derived from the CW system and the run of the Excel optimisation programme, the values of the constants K_1 and K_2 were determined to be 380.7 and 154.8 h^{-1} (using the dataset for BOD_5) and 243.4 and 199.3 per hour (using the dataset for COD). These values for both BOD_5 and COD were then separately used in Eq. 7.9 to obtain the mathematical model. The results obtained from testing the mathematical model are presented in Figs 7.1 and 7.2. Figure 7.1 shows the plot of the predicted and observed experimental values for each respective influent value of COD and BOD_5, while Fig. 7.2 shows the plot of the PD for each influent value. From Fig. 7.1, the mathematical model can be described as having a 'moderate' fit with the experimental data. The PD was much higher for BOD_5 than for COD. Nonetheless, it can be seen that the mathematical model still needs further investigation. The values of the constants K_1 and K_2 obtained from the model optimisation using the dataset of this study were higher by 3 to 4 orders of magnitude than the respective values obtained by Sun et al. (1998).

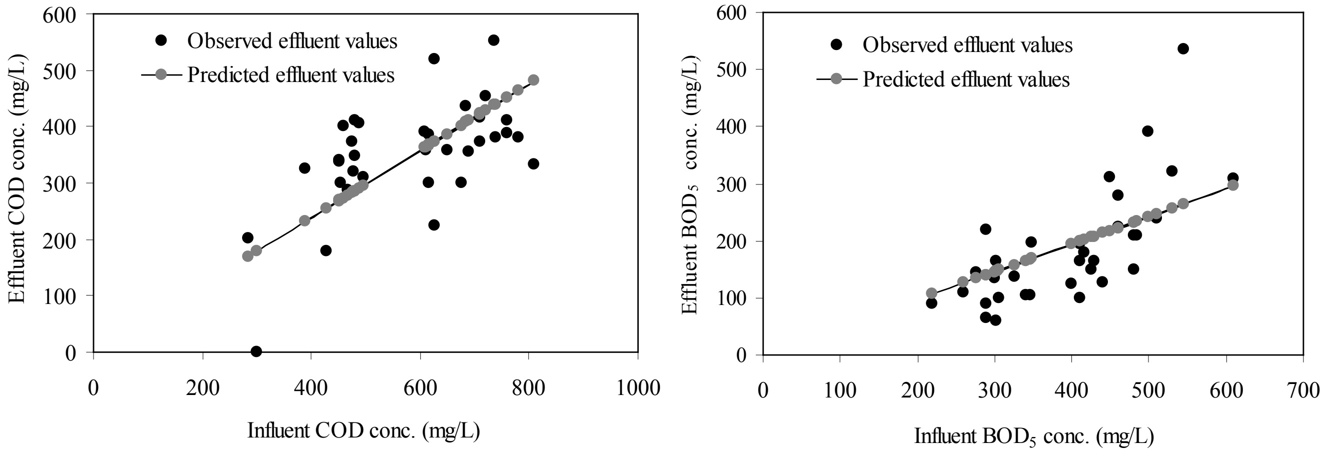


Figure 7.1. Predicted and observed COD and BOD_5 effluent concentration values plotted for each respective influent concentration value.

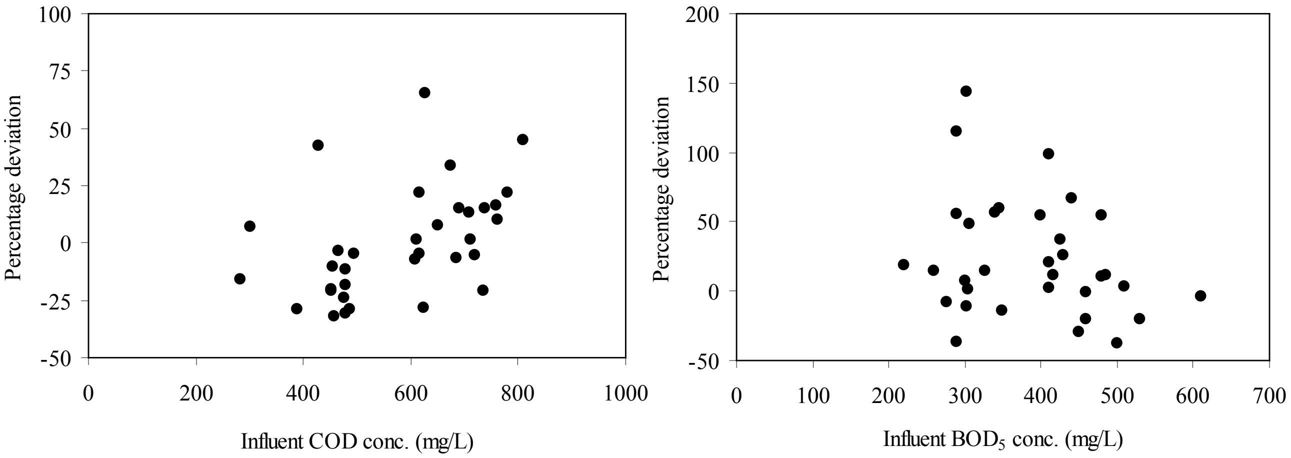


Figure 7.2. Plot of percentage deviation based on model prediction for each corresponding influent concentration value for COD and BOD$_5$.

While the relatively high optimal value obtained for K_1 might possibly be explained by the high adsorption capacity of the alum sludge, the high value obtained for K_2 might suggest the contribution and influence of processes other than biological degradation to the removal of organic matter. Indeed it is recommended that the model should be used for wastewaters with BOD$_5$ greater than 200 mg/L, and in particular when other processes such as plant uptake and filtration do not have a pronounced influence on the treatment result (Sun et al., 1998).

7.3 Summary

Results presented in this section have demonstrated the potential feasibility of using a mathematical model to describe the treatment behaviour in the novel alum sludge-based CW system. Results show that the model can be described as producing only a moderate fit between the predicted and the observed values for COD and BOD$_5$. Further study in this respect is recommended.

8 Making Alum Sludge an Environmentally Safe Material for Reuse

During the different stages of study on the development of alum sludge-based CW for wastewater treatment, alum sludge was investigated as a main substrate in various models of CW system for *P*-rich wastewater treatment. The alum sludge-based CW is a promising technology with a two-prong feature of 'waste' reuse and wastewater treatment. However, it is necessary to address some relevant concerns pertaining to the safety and practicality of its reuse such as:

1. Is alum sludge suitable for reuse in a CW?
2. Is Al release a concern?
3. How can *P* be recovered from the alum sludge?
4. How can polymer residual be monitored in the alum sludge?

8.1 Is Alum Sludge Suitable for Reuse in a Constructed Wetland?

Since alum sludge is derived from the residual of the treatment of raw water, which contains mainly turbidity, colour, suspended clays and humic substances, it is unlikely to contain a substantial quantity of toxic substances. The alum sludge used in this project was characterised to assess its suitability as a substrate in a CW.

8.1.1 *Examination of Physico-Chemical Characteristics*

The porosity was determined from the amount of water needed to saturate a known volume of the sludge while bulk density was determined from the volume of water displaced by a known mass of the sludge sample (*n* = 5). The surface area was measured using nitrogen Brunauer–Emmett–Teller (BET) isotherms on a Micromeritics Gemini 2375 volumetric analyser. The total pore volume was calculated from the amount of vapour absorbed at a relative pressure (P^o/P, where P is the equilibrium pressure and P^o is the saturation pressure of the adsorbate) close to unity. The micropore volume of the alum sludge was calculated using the Dubinin–Radushkevich (DR) model of micropore filling, which is based on Polanyi's concept of a characteristic curve to describe adsorption into micropores (Gregg and Sing, 1982). The characteristic curve can be expressed as in Eq. 8.1:

$$\frac{V}{V^o} = \exp\left(-\frac{A}{\varepsilon\beta}\right)^2 \qquad (8.1)$$

Where V is the volume adsorbed at the relative pressure, V^o is the micropore volume, A is the affinity or differential free energy of adsorption [$A = RT\ln\left(\frac{P^o}{P}\right)$], ε is the characteristic free energy of adsorption for a given system (equals A when $V/V^o = 1/e = 0.368$, where e is the base of natural logarithm) and β is the similarity factor. Substituting for the value of A in Eq. 8.1 and taking logarithm gives the known DR equation as Eq. 8.2:

$$\log V = \log V^o - D\log^2\left[\frac{P^o}{P}\right] \qquad (8.2)$$

Where D is a constant characteristic of the adsorbent structure related to the characteristic energy according to Eq. 8.3:

$$D = \frac{R^2T^2\ln 10}{\varepsilon^2\beta^2} \qquad (8.3)$$

As a first approximation, the micropore volume can be determined directly from the BET-N_2 adsorption isotherm, and it corresponds to the volume of gas adsorbed at P/P^o of 0.15 (Gregg, 1982). However, from Eq. 8.2, a graph of log V against $\log^2(P^o/P)$ will give the micropore volume of the alum sludge, V^o, as the intercept when extrapolated from the linear portion of the DR plot, assuming that the pores are filled with the liquid adsorbent (Echeveria, 1999). The particle size distribution (PSD) of the alum sludge was examined using standard sieve analysis technique and the values of d_{10} and d_{60} and the uniformity coefficient (UC) were determined. Surface morphology and microstructure were examined using scanning electron microscopy (SEM (JEOL JSM 5510) to visualise inner porosity, surface properties and a potential environments for biofilm bacteria. Roger (2000) noted that biofilm bacteria need a potential environment of typically 1 to 3 µm. The SEM was further combined with EDX (energy dispersive X-ray, INCAx-Sight model 6587, Oxford Instruments, UK) to determine the composition and relative distribution of elements, particularly on the alum sludge surface. X-ray diffraction of randomly oriented powders of the samples was carried out on a Phillips PW 1050 diffractometer using monochromated CuK_α radiation, operating at 40 kV and 30 mA, from 3° to 75° 2Θ, with a scanning step of 0.02° at 1° per minute while the Fourier transform infrared (FTIR) spectra was measured using the KBr wafer technique in a FTIR spectrometer (Excalibur series).

The chemical properties were also determined as follows: pH was taken as the pH of a 0.01 M $CaCl_2$ solution of the alum sludge (McLean, 1982) (particle size <2 mm, alum sludge: solution ratio of 1:10). Loss of ignition (LOI) was determined in accordance with standards (British Standards Institution, 1990). Electrical conductivity (EC) was measured at 25°C using a Radiometer CDM83 electrical conductivity meter (Smith and Doran, 1996). The elemental metal composition was carried out by carefully weighing ca. 0.025 g of the air-dried alum sludge samples (particle size <2 mm) into clean trifluoromethylene (TMF) vessels followed by the addition of 4 ml HNO_3 + 200 µl HF + 4 ml H_2O. Samples were then digested using a low-volume microwave digestion technique (Sandroni et al., 2003), and the digestates analysed for dissolved metals using inductively coupled plasma mass spectrometry (ICP-MS). Specific anions (Cl^-, SO_4^{2-}, SiO_4^{2-}) and humic acid (expressed as TOC) were determined separately (Yang et al., 2006b).

8.1.2 Results

The characterisation results show that the alum sludge had a bulk density of 1.18±0.11 g cm^{-3} and a porosity of 45%, both of which are comparable to values of 0.7 to 1.83 g cm^{-3} (bulk density) and 30 to 54.4% (porosity) reported in the literature for potential candidate materials for CW (Drizo et al., 1999; Roger, 2000; Del Bubba et al., 2003). From the PSD, the values of d_{10}, d_{60} and UC were computed to be 0.5, 1.8 and 3.6 respectively, and these are consistent with several guidelines for securing adequate hydraulic conductivity and minimising the risk of clogging (IWA, 2000). The microstructure of the alum sludge (Fig. 8.1 [a&b]) suggests the abundance of Al ions on the surface of the alum sludge and a 'rough' surface

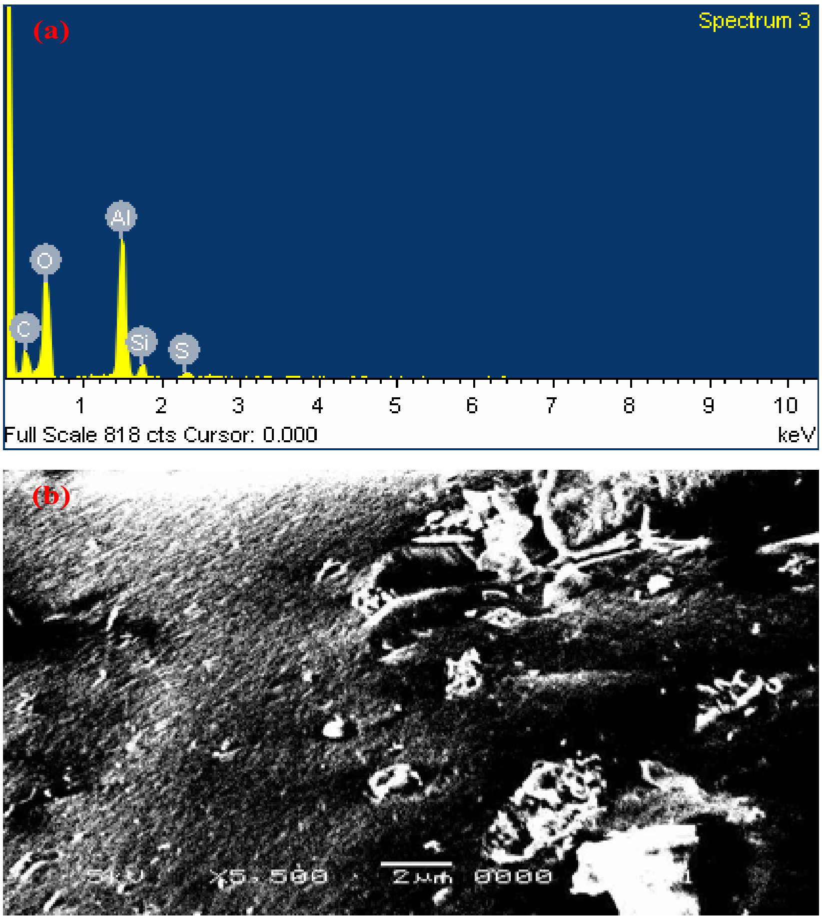


Figure 8.1. Surface images of the alum sludge using (a) SEM-EDX and (b) SEM.

that would be ideal for biofilm growth. The specific surface area (SSA) ranged from 28.0 to 41.4 m^2/g. Makris et al. (2004) found that alum sludges can have an SSA up to 105 m^2/g. Furthermore, the SSA of the alum sludge was found to increase with increasing particle size, suggesting a large inner porosity in the grains. In comparison to the range of SSA of other candidate wetland materials reported in the literature, such as 2.6 to 3.9 m^2/g (Roger, 2000) and 6.8 to 31.4 m^2/g (Drizo et al., 1999), the alum sludge used herein can be seen to have a comparatively higher SSA. This further relates to having an adequate surface area for biofilm growth and attachment. The micropore, mesopore and pore volumes of the alum sludge were determined to be 0.0125 cm^3/g, 0.0210 $cm^3 g^{-1}$ and 0.0410 cm^3/g respectively. The mesopore volume was ca. 1.7 times the micropore volume and the mesopores also account for about 51.2% of the entire pore volume of the alum sludge. The X-Ray diffraction pattern did not reveal any sharp diffraction characteristic peak over a broad range of d-spacings (10 to 80°, 2Θ). This indicates poorly ordered particles within the alum sludge and it also illustrates that, even though the SEM-EDX results highlight the predominance of Al in the alum sludge, it is likely to be amorphous Al. This observation is also in agreement with other authors (Dayton and Basta, 2001; Makris and O'Connor, 2007). Similarly, the FTIR spectrum ranging from 500 to 4000/cm did not provide clear information regarding peaks.

The EC of the alum sludge ranged from 0.104 to 0.140 dS/m, and thus the sludge can be considered non-saline (Smith and Doran, 1996). The EC of the sludge is also well below the 4 dS/m associated with reduced plant growth due to salinity (Dayton and Basta, 2001). It is important to note that microbial mediated processes, which are the basis of wetland operations, are very sensitive to soil EC, hence the importance of EC. The alum sludge had a residual pH in the range of 5.9 to 6.0, which was within a typical pH range of 5.1 to 8.0 for water-treatment residuals reported by Dayton and Basta (2001). The TOC of the alum sludge averaged 97.5 mg/g (Yang et al., 2006b), while the LOI averaged 49.4%. These values may be attributed to humic substances contained in the raw water being treated. The elemental composition of the alum sludge, in comparison with some other wetland materials, is given in Table 8.1. It is clear that the primary coagulant (aluminium sulphate) used during the water-treatment process is reflected in the composition of the sludge. The Al component in the alum sludge is about 1 to 5 orders of magnitude greater than all other elements analysed in the alum sludge.

Table 8.1. Major elemental chemical composition of the Al-based water-treatment residuals compared to other water-treatment residuals and potential wetland media.

Element (mg/g)	Water treatment residual				Other potential media	
	a	b	c	d	e	f
Al	42.67	38.3-125.4	15-300	297	nd	0.065 – 0.084
Fe	3.336	16.3–26.1	5-66	102	nd	21.9–45. 8
Ca	0.820	4.5–54.6	3-50	29	nd	12.1–34
P	0.123	0.5–4.4	0.2–4.4	3.5	nd	nd
As	0.034	0.007–0.04	nd	nd	0.00–0.07	nd
Pb	0.005	0.005–0.032	nd	0.04	0.03–11.69	nd
Mg	0.237	0.054–0.142	nd	8.9	nd	6.5–27.1
Mn	0.270	0.036–2.688	nd	nd	nd	0.39–0.846
Ti	0.099	nd	nd	nd	nd	nd
Zn	0.03	0.05–0.0017	nd	0.03	0.07–7.89	nd
*Cl^-	16.1	nd	nd	nd	nd	nd
*SO_4^{2-}	8.3	nd	nd	nd	nd	nd
*SiO_4^{2-}	11.2	nd	nd	nd	nd	nd
*TOC	97.5	nd	8.5-225**	nd	nd	nd

[a]This study, [b] DeWolfe (2006), [c]Makris and O'Connor, (2007), [d]Babatunde and Zhao (2007), [e]Page et al. (2003), [f]Kvarnstrom, et al. (2004), *mean values obtained from Yang et al. (2006b), nd: no data, **total carbon. Determinations were carried out on an air-dried Al-based water-treatment residuals with particle sizes <2mm.

The Al content is also quite high when compared with some other potential wetland filters as reported by Kvarnstrom et al. (2004), but the calcium and iron levels in the alum sludge are comparatively low. Table 8.1 also shows that the concentration of the major constituents in the alum sludge are within the typical ranges as reported in literature for water-treatment sludges (DeWolfe, 2006; Babatunde and Zhao, 2007; Makris and O'Connor, 2007), although a wide variability amongst samples is observed. The arsenic concentration in the alum sludge is however much greater than the mean value of 0.0113 mg/g reported for five alum sludges as reported by Makris and O'Connor (2007), while levels of lead and zinc in the alum sludge are below their typical levels in uncontaminated soils.

8.1.3 Summary

The results demonstrated that alum sludge has an ideal surface for biofilm growth and attachment with an SSA ranging from 28.0 m^2/g to 41.4 m^2/g. Both the pH and the EC of the sludge showed that it should support plant growth suitably, while chemical analysis also indicates that there is no component of the sludge that should significantly preclude its use as a substrate in CWs. More importantly, compared with the commonly used substrates in CWs such as sand, gravel and local soil, the alum sludge from the Ballymore-Eustace water treatment plant is comparable in its characteristics and it can be used in CWs, especially for P-rich wastewater treatment.

8.2 Is Aluminium Release a Concern?

One of the concerns of the beneficial reuse of dewatered alum sludge in CWs is the possible release of Al from the sludge since the sludge is enriched in Al. Accordingly, in this study, a brief review of the release of Al during the reuse of the alum sludge was conducted and a 42-week Al monitoring was carried out on the field CW to monitor potential Al release.

8.2.1 Aluminium Concentration in Regulations

Several regulations have been promulgated in relation to Al concentrations in drinking waters and effluents for discharge. In Ireland and the UK, the prescribed limit for Al discharge into all waters is 0.2 mg/L (Council Directive 98/83/EC, 1998). For drinking waters, the World Health Organization suggests a maximum limit of 0.2 mg/L (World Health Organization, 1998) while in the USA, the US Environmental Protection Agency secondary drinking water regulation stipulates a range of 0.05 to 0.2 mg/L (Kvech and Edwards, 2002).

8.2.2 Brief Review on Aluminium Release from Studies Focusing on Alum Sludge Reuse

A preliminary screening of available information and published work on the use of alum sludge for P removal and its application as a substrate in CWs was conducted. Results show that except for the studies where Al release was not particularly monitored or studied, all the other studies reported observed Al release. Out of the 35 studies reviewed, Al release was mentioned or observed in 22 studies. However, although Al release was not reported in 13 of the studies, it cannot be concluded in these cases whether Al was released or not. The concentration of leached/soluble Al reported ranged from 0.01 to about 20 mg/L and the influence of pH was significant. However, except for three reported values of 1.11 mg/L, 7 mg/L (pH 3) and about 20 mg/L (pH 3), all other values reported were <1 mg/L and fell mostly between <0.01 and <0.2 mg/L. In terms of pH, Al release was found to be most significant at pH 3 with values of ~7 mg/L (Hovsepyan and Bonzongo, 2008) and >20 mg/L (Mortula et al., 2007a) reported. Furthermore, the review shows that higher Al leachate values were observed at the beginning of the experiments in some cases, but this decreased over the course of the experiment. The impact of Al release was evaluated, in some cases with mixed conclusions obtained. For instance, Mortula et al. (2007b) reported Al leaching in their study but concluded that the Al level in the leachate was within a reasonable range for surface water disposal and the level is not high enough to cause toxicity for aquatic species if disposed. Agyin-Birikorang et al. (2009) also concluded in their study that alum sludge can be safely used to reduce P leaching into groundwater without increasing the aluminium concentration of the groundwater. However, although Kaggwa et al. (2001) in a study on the discharge of alum sludge to a natural wetland reported that there was no adverse effect on the water quality and the growth and development of the aerial biomass, Al toxicity in the rooting system was reported to cause root abnormalities.

8.2.3 *Monitoring of Al from the Field Pilot-Scale Alum Sludge-Based CW System*

8.2.3.1 The field pilot-scale CW set up and Al monitoring

The field pilot-scale alum sludge-based CW system has been described in detail in Section 7.1. Animal farm wastewater was treated in the novel CW system. Samples of influent and effluent were collected weekly for the first 4 weeks and every 2 weeks thereafter (except for the period between the 12th and 18th weeks when the system was stopped to allow for some changes on the farm) and analysed for total and soluble Al.

8.2.3.2 Results of Al Monitoring

Levels of total and soluble Al monitored in the influent and effluents of the individual stages of the CW system during the operation are shown in Fig. 8.2.

This shows that the levels of total and soluble Al were mostly higher in the effluents than in the influent and this indicates a release of Al. However, it can be observed from the figure that the highest levels of total and soluble Al were detected during the first 3 weeks of operation and the levels decreased afterwards, except for the 26th week when a rise in both parameters was observed. Beyond the first 3 weeks, concentrations of

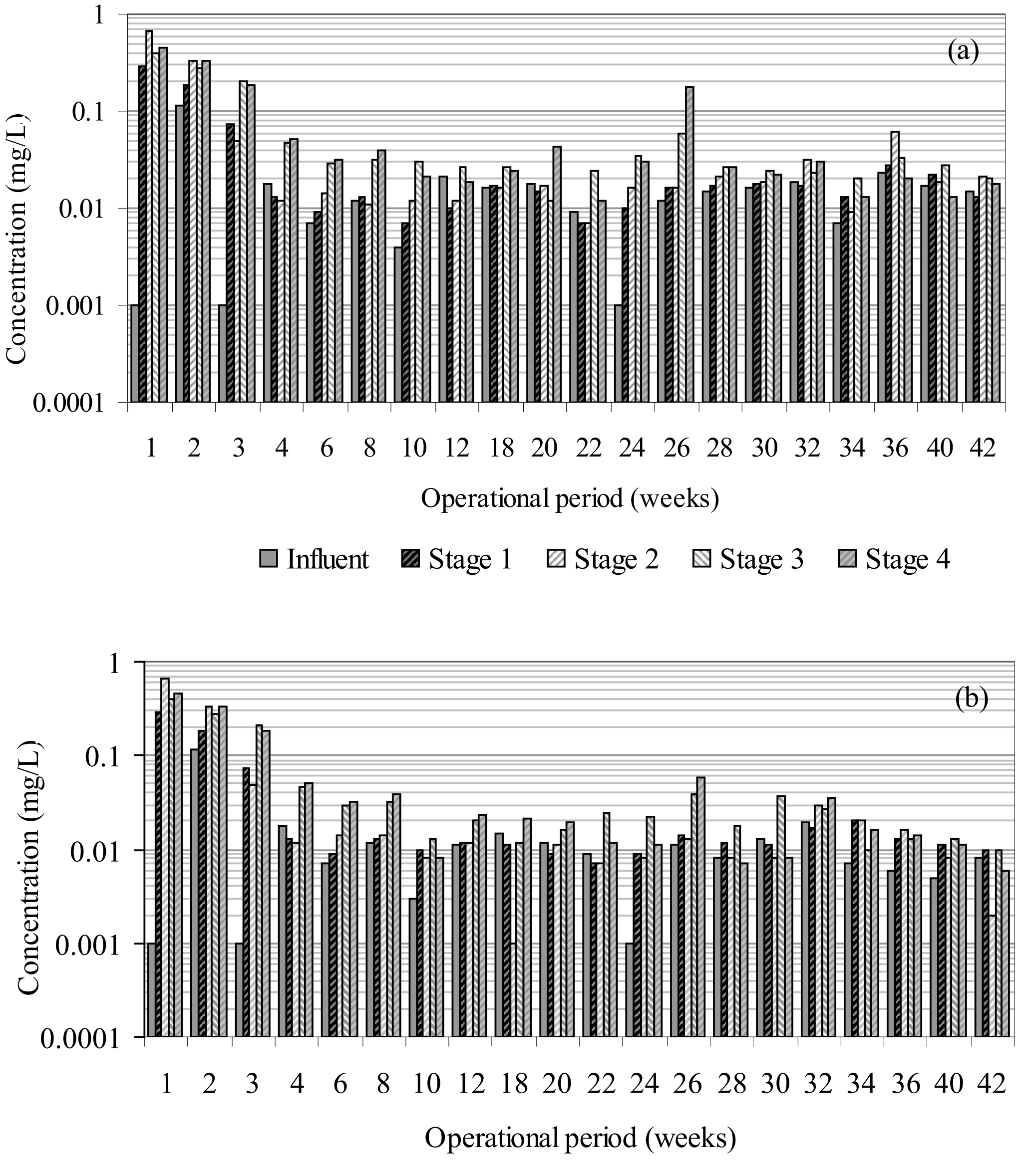


Figure 8.2. Monitoring results for aluminium concentration in the influents and effluents of the constructed wetland system showing (a) total Al and (b) soluble Al.

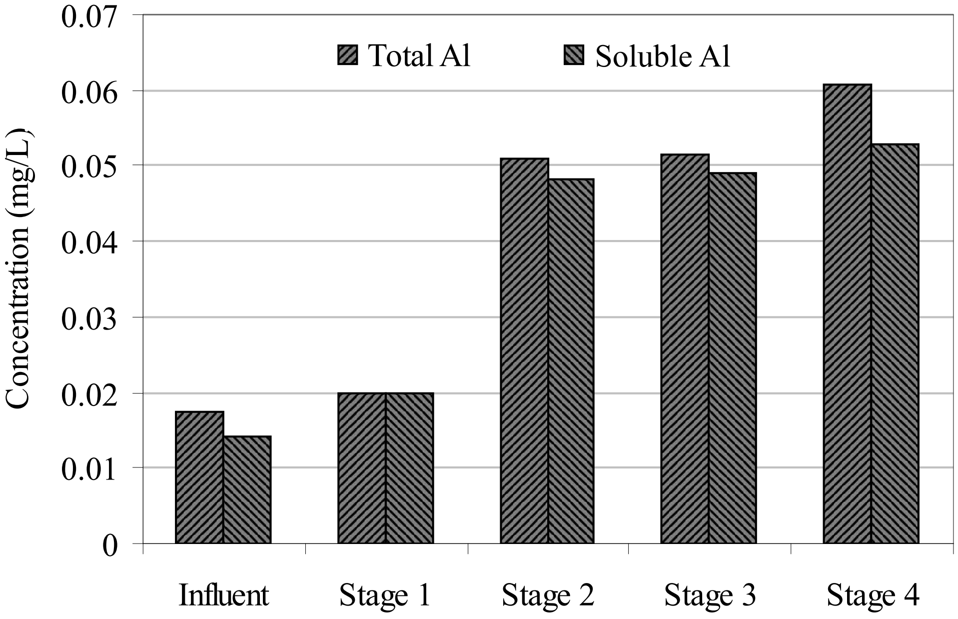


Figure 8.3. Mean levels of influent and leached aluminium in the different stages of the constructed wetland system.

total and soluble Al in the effluents remained below the recommended guideline limit of 0.2 mg/L for drinking water standard and effluent discharge. Al was also detected in the influent wastewater into the CW. By accounting for this background Al concentration in the wastewater being treated, the level of Al leached in each stage of the CW was determined (see Fig. 8.3). The levels of Al leached were generally low and range from 0.02 to 0.06 mg/L across the stages, with leached level of Al increasing across the stages.

Figure 8.4 shows the pattern of P removal and Al release in the individual stages of the CW. An inverse relationship was observed but regression analyses indicate that the relationship is weak. It can also be seen that whereas P concentration decreased from the influent and across the stages, both total and soluble Al showed an opposite trend, with their concentrations increasing from the influent and across the stages. It therefore follows that as more P is being removed,

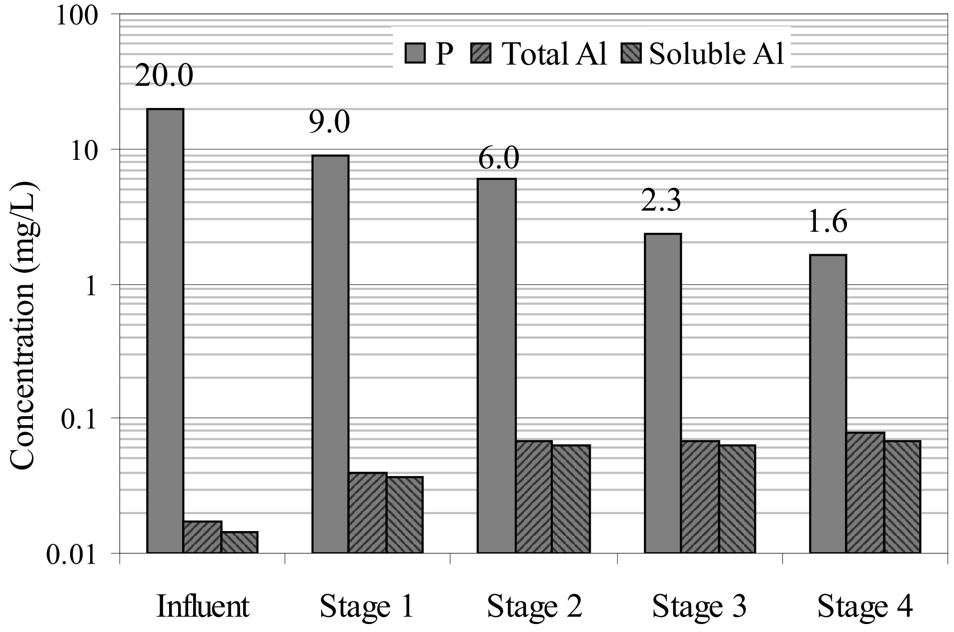


Figure 8.4. Trend of phosphorus reduction and release of aluminium from the influent and across the stages of the constructed wetland system (mean values plotted).

mainly through adsorption on the alum sludge, the tendency for the leaching out of Al is reduced.

8.2.3.3 Discussion

The results obtained in this study show that the level of Al leached was quite low, ranging from 0.02 to 0.06 mg/L and the overall concentration of Al in the effluents were well below regulatory guideline limits of 0.2 mg/L except for the first 3 weeks of operation. It is noted that the wastewater being treated had some background Al concentration and this may have contributed to the overall concentration of Al in the effluents.

It is therefore very important to consider the background Al concentration of the wastewater being treated to prevent an exaggerated view of Al release from the alum sludge during such reuse. However, even though the levels of Al found in the effluents do not represent an imminent environmental or health risk, periodic determinations are advisable. In a previous study of four laboratory-scale CWs using alum sludge as a substrate in different proportions (Section 3), it was found that the concentrations of total and soluble (dissolved) Al in the effluent were above the prescribed limits for discharge in most cases – especially at the beginning of the experiments. In particular, levels of soluble Al monitored in the effluents was reported to range from 0.058 mg/L to 1.106 mg/L. However, the wastewater being treated in the study had some background Al concentration, which would have contributed to the levels of Al observed.

On the effect of P adsorption on Al release, the result obtained in this study indicates that P adsorption onto the alum sludge may have served to reduce Al leaching from the sludge. An analysis of P removed and Al released in each stage of the CW reveals an inverse trend between P adsorbed and Al released across the four stages of the CW. It therefore follows that P adsorption onto the alum sludge may contribute to reducing Al leaching from the alum sludge, but it is not a direct relationship. Mortula et al. (2007a) reported similar findings in their studies that P adsorption on alum sludge may have an insignificant effect on Al release.

8.2.4 Summary

Regarding the concerns about Al release from the reuse of alum sludge, a review of literature shows that most of the related studies were laboratory based and 22 out of the 35 studies mentioned/reported Al release. Concentration of leached/soluble Al reported ranged from 0.01 to about 20 mg/L and the influence of pH was noted. However, except for three reported values, which ranged from 1.11 to 20 mg/L, all other values reported were <1 mg/L and mostly between <0.01 and <0.2 mg/L. Monitoring of Al in the field-scale CW shows that there was Al leaching. However, except for the first 3 weeks of operation, effluents concentrations of both total and soluble Al were all below the limit of 0.2 mg/L set as a drinking water quality standard.

8.3 Phosphorus Recovery from Saturated Alum Sludge in Constructed Wetlands

It has been demonstrated that over 90% of P can be absorbed/removed by a dewatered alum sludge-based CW system. This converts the sludge from a ‘waste’ into potentially useful raw material. However, there are concerns as regards the final fate of the P-saturated sludge after reuse in CW. Therefore, the objective of this phase of study was to investigate the feasibility and effectiveness of P recovery from saturated alum sludge that was used as substrate in CW for P immobilisation. A P-recovery process involving P desorption, the Fenton reagent reaction for decolouration of the P-extraction leachate, followed by aluminium phosphate formation by pH adjustment of the decolourised leachate was developed.

8.3.1 Materials and Methods

8.3.1.1 Materials

The P-saturated alum sludge used in this study was obtained from the laboratory-scale CW treating P-rich real animal farm wastewater. After a long-term operation, the alum sludge was almost saturated with P. The sludge was then dried at room temperature and ground and sieved to a diameter of <0.3 mm to provide the test samples. The sludge samples, both fresh (unused) and P-saturated (used), have been characterised (Zhao and Zhao, 2009) and the main chemical components are listed in Table 8.2. Analytical grade hydrogen peroxide solution (30% w/w) and ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$) were purchased from the Riedel-deHaën Chemical Company and were used as received.

Table 8.2. Chemical components of the alum sludge.

Chemical composition	Unit	Fresh alum sludge	P-saturated alum sludge
Total organic carbon (TOC)	%	12.8	6.9
Aluminium (Al)	mg/kg	42,880	63,020
Calcium (Ca)	mg/kg	1964	16,460
Iron (Fe)	mg/kg	2465	3758
Magnesium (Mg)	mg/kg	46	2377
Phosphorus (P)	mg/kg	154	38,590
Nitrate as NO_3	mg/kg	42	366
Nitrite as NO_2	mg/kg	0.4	3.2

8.3.1.2 *Experimental Procedure*

In P-desorption tests, the efficiencies of acid and base solutions with different concentrations for phosphorus extraction in 60 min were investigated. A batch of 0.8 g pre-weighed sludge samples was put into a series of 250 ml flasks. Then, 150 ml of acid (HCl, HNO_3 or H_2SO_4) or base (NaOH or KOH) with different H+/OH^- concentrations up to 0.1 M were added to the sample. Thereafter, the flasks were placed on an orbital shaker (SSL1, Bibby Sterilin Ltd, UK) at 225 rpm for 1 hour for P release. After P extraction, the sampling was filtered using 0.45 µm Millipore filter paper and diluted before the analysis of phosphate (PO_4-P). A Hach spectrophotometer (DR/2400) was used for P analysis according to the standard method. Following the P extraction, the leachate was a dark-red solution. The second set of experiments was conducted to examine the efficiency of Fenton reagent for decolouration of H_2SO_4 leachate. 100 ml of the solution was put into a 250 ml beaker and different quantities of hydrogen peroxide (H_2O_2) solution and ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$) were added. Thereafter, the solution was stirred gently and samples were taken from the beaker every hour for colour testing. Finally, a series of 100 ml decolourised H_2SO_4 leachate was adjusted using 4 M NaOH at pH range of 4 to 10.5 and stirred for 30 min. Thereafter, the precipitates were centrifuged at 3500 rpm for 10 minutes and dried at 80°C. The supernatants were then analysed for P and Al residual concentration.

8.3.2 *Results and Discussion*

8.3.2.1 *Phosphorus Release by Acid and Base Extraction*

The P adsorption by alum sludge is dominated by the significant amount of reactive functional groups, such as –OH, –Cl, $–SO_4$ and humic substances in the sludge surface through the ligand exchange mechanism. Due to the significant amount of Al in the sludge, P and Al can form complex compounds through chemical reaction and precipitation (Yang et al., 2006b). Accordingly, the main component can be simply described as alum phosphate. Therefore, the leaching reaction with acid or base can be expressed as the chemical equations Eqs 8.4 and 8.5:

$$AlPO_4 + 3H+ \rightarrow Al_3+ + H_3PO_4 \qquad (8.4)$$

$$AlPO_4 + 4OH_- \rightarrow Al(OH)_{4-} + PO_4^{3-} \qquad (8.5)$$

Table 8.3 illustrates P release efficiency by different acids (HCl, HNO_3, H_2SO_4) or bases (NaOH, KOH) with different concentrations at 60 minutes. From the results, there is no obvious difference in the extraction abilities between the different acids or bases and the extraction efficiencies exhibited an increasing trend with concentrations of H^+ or OH^-, but there was no significant increase observed between 0.075 and 0.1 M. When $AlPO_4$ is dissolved by acid or base, it reacts with H^+ or OH^- (see Eqs 8.4 and 8.5 above). It is interesting to note that in low concentrations under 0.05 M P release was higher for the base than the acid, but in high concentrations, the reverse is the case.

Table 8.3. Phosphorus release efficiency by different acid or base with different H^+/OH^- concentrations.

	HCl	HNO_3	H_2SO^4	NaOH	KOH
0.01 M	4.2	4.2	4.8	16.3	17.8
0.03 M	41.2	42.0	37.5	46.0	52.5
0.05 M	75.0	72.2	72.3	60.8	61.2
0.075 M	85.8	89.8	88.5	65.5	63.3
0.1 M	90.7	91.0	88.5	66.1	65.3

Unit: %.

For example, the P released by 0.075 M HNO_3 and NaOH were 90% and 65% respectively. The reason may be due to the $Al(OH)_3$ inside the alum sludge. At low concentrations, the residual $Al(OH)_3$ would consume some acid or base as described in Eqs 8.6 and 8.7:

$$Al(OH)_3 + 3H^+ \rightarrow Al^{3+} + 3H_2O \quad (8.6)$$

$$Al(OH)_3 + OH^- \rightarrow AlO_2^- + 2H_2O \quad (8.7)$$

Obviously, more acid was used for dissolving $Al(OH)_3$. Thus, the acids showed lower efficiency for P release than the bases. But when concentrations are high enough, the reactions mainly follow Eqs 8.4 and 8.5. By considering the cost of HNO_3, HCl and H_2SO_4 for the reagents for P extraction, H_2SO_4 was chosen for the subsequent experiments since H_2SO_4 extraction is much cheaper than HNO_3 and HCl. In addition, considering the convenience and safety of the operation, H_2SO_4 is better than the others because HNO_3 and HCl are both highly volatile acids with dangerous acid mists. The optimal condition for H_2SO_4 extraction has been studied using the response surface methodology and the best P extraction efficiency of 98% can been achieved at the condition of: (i) mass of sludge of 0.8g, (ii) H_2SO_4 concentration of 0.063 M and (iii) volume of H_2SO_4 solution of 142 ml (Zhao and Zhao, 2009).

8.3.2.2 Decolouration of H_2SO_4 Leachate by Fenton Treatment

The H_2SO_4 leachate for P extraction was a dark-red solution with a P concentration of 720.8 mg-PO_4^{3}/L under the optimal extraction conditions. By using ICP-MS, a Hach spectrophotometer and a pH meter, the characteristics of the solution were determined, and the main parameters were: pH (2.1), colour (4100 Pt-Co units), COD (1026 mg/L), TOC (430 mg/L), Al (346.4 mg/L), Ca (74.8 mg/L), Mg (10.9 mg/L) and Fe (9.0 mg/L). In order to recover the pure phosphate, decolourising of the H_2SO_4 leachate will be necessary. The Fenton reagent, consisting of H_2O_2 and ferrous iron (Fe^{2+}), has been shown to be effective in the degradation of organic and inorganic pollutants (Neyens and Baeyens, 2003; Pignatello et al., 2006). The Fenton reaction involves the formation of HO• radicals as the actual oxidants (Sanly et al., 2007, Kochany et al., 2008). Decolouration and degradation using Fenton reagent were conducted and the results are illustrated in Fig. 8.5. As expected, the colour removal increased with increasing concentration of Fe^{2+} addition within the reaction period of 8 hours. This

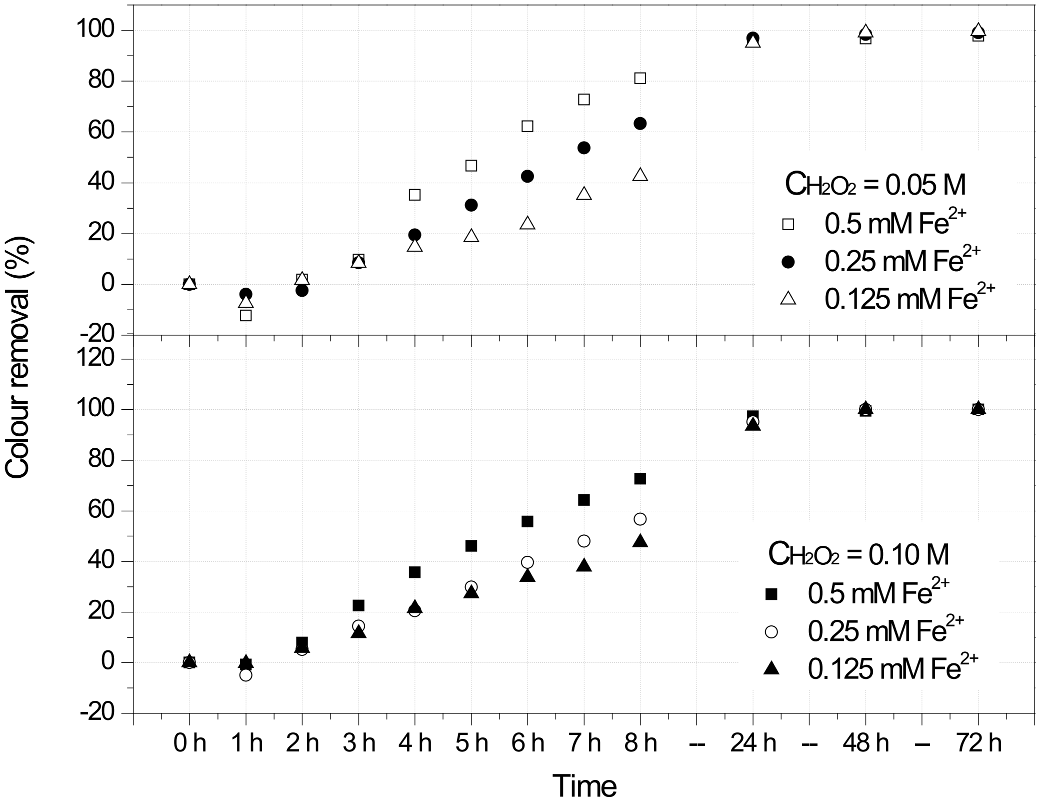


Figure 8.5. Colour removal at different H_2O_2 and Fe^{2+} dosages.

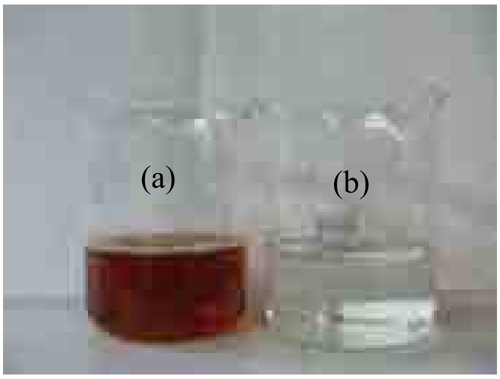


Figure 8.6. Photograph of H_2SO_4 leachate (a) before and (b) after Fenton treatment.

is because the production rate of hydroxyl radical (HO•) has a linear relation with concentrations of Fe^{2+} in the first stage of the Fenton process (Gao et al., 2006). This can also be confirmed by the decrease of H_2O_2. High Fe^{2+} concentrations can promote the Fenton reaction. Interestingly, it is noted that, when Fe^{2+} concentrations remain at the same levels, adding 0.1 M H_2O_2 gives less colour removal efficiency than that of 0.05 M over 8 hours. This is because of the self-decomposition of H_2O_2 to O_2 and H_2O, and the recombination of HO• radicals (Buxton et al., 1988). However, it can be seen from Fig. 8.5 that, after 24 hours' reaction, both 0.05 M and 0.1 M H_2O_2 showed a similar ability for colour removal. It should be pointed out that in the final stage of the Fenton reaction higher Fe^{2+} concentrations would have a negative effect on colour removal.

8.3.2.3 Phosphorus Recovery by Deposition

By inspecting the data of Al and PO_4^{3-} concentrations in the H_2SO_4 leachate, it can be seen that the molar ratio of Al^{3+}/ PO_4^{3-} is suitable to form $AlPO_4$ by deposition. Therefore, through adjusting the pH of the solution by adding NaOH, the precipitates were formed, as shown in Fig. 8.7(a). The precipitate after centrifugation is shown in Fig. 8.7(b). The results of P deposition efficiency by pH adjustment of the decoloured H_2SO_4 leachate are illustrated in Fig. 8.8. The figure indicates that the pH is the most important factor for $AlPO_4$ formation. The suitable pH range is 5 to 7, at which over 98% P can be deposited. Recovered phosphate, which is mainly composed of aluminium phosphate, has several potential uses, such as serving as an absorbent, as raw material in industry, and as a fertiliser in agriculture, etc.

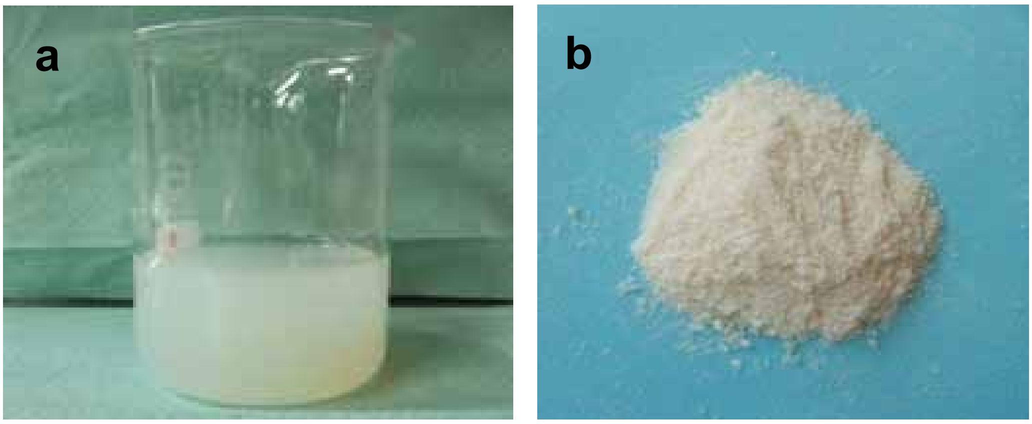


Figure 8.7. White sediment ($AlPO_4$) in solution (a) after deposition reaction and (b) after centrifugation.

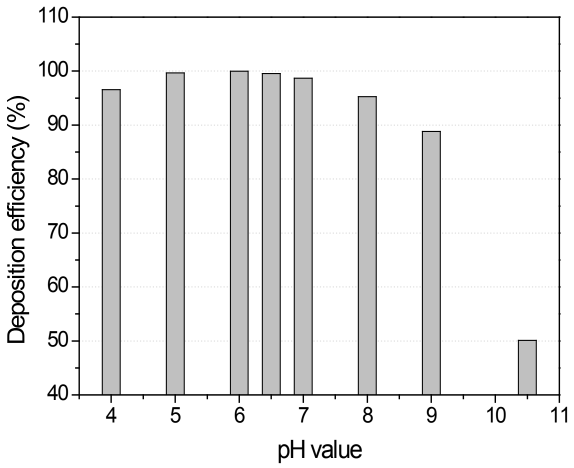


Figure 8.8. Mean values of P deposition efficiency under different pH at reaction of 30 min.

8.3.3 Summary

A P-recovery process, which includes H_2SO_4 extraction, Fenton (H_2O_2/Fe^{2+}) reaction for decolouration and pH adjustment of the decoloured H_2SO_4 leachate, was investigated. The use of different acids (HCl, HNO_3, H_2SO_4) and bases (NaOH, KOH) as P-extraction reagents was examined. The results indicated that both acid and base were efficient for P extraction and the efficiency relied mainly on the concentrations of H^+/OH^- and not the type of acid or base. H_2SO_4 was then chosen as the most suitable reagent for P extraction. Fenton treatment was demonstrated to be an efficient method not only for colour but also for COD and TOC removal from the H_2SO_4 leachate. By adjusting the pH of decolorised H_2SO_4 leachate to 6, the white precipitate of $AlPO_4$ was formed, which can be possibly used as raw material and fertiliser in industry and agriculture.

8.4 Polymer Monitoring and its Possible Release from the Alum Sludge

Polymers or polyelectrolytes consist of simple monomers that are polymerised into high-molecular-weight substances with molecular weights varying from 104 to 106 Daltons. Although such organic polymers are specifically manufactured for the water industry, the monomers used in the manufacture of many polyelectrolytes are toxic. Their global usage, particularly as a flocculant aid in conjunction with a metal salt coagulant during drinking water treatment processes, and also in sludge conditioning/flocculation, has raised increased concerns about the impacts of the polymer residuals in the surrounding environment when the dewatered sludge is recycled/reused or landfilled. Specific long-term effects of the polymer residual on aquatic organisms, human health and the biota when the sludge is disposed of are still relatively unknown. In this phase of study, the methodology of direct measurement of polymer residual using high performance liquid chromatography (HPLC) was developed. Thereafter, the investigation of polymer residual in the alum sludge reused in the CW was investigated on a preliminarily basis. An overview of the potential problem regarding polymer residual in the water environmental is presented.

8.4.1 Concerns of Organic Polymers: Mini Review

A review of polymers used in the water industry was given by Bolto and Gregory (2007). Unfortunately, techniques for appropriate monitoring of the polymer residual are lacking (Zhao et al., 2008b). This implies that the polymer residual in the water treatment and sludge conditioning processes is substantially unknown although the amount of the polymer addition is accurately known. Furthermore, information on the fate of the added polymers in water and sludge treatment is lacking. It has been noted that the use of polyelectrolytes in Japan and Switzerland is not permitted in the treatment of drinking water, while Germany and France have set a strict limit for such use (Bolto and Gregory, 2007). Therefore, new concepts and principles for sludge conditioning/treatment in line with sustainable development remain a significant challenge to engineers and scientists.

8.4.2 Development of a Methodology for Polymer Residual Monitoring using HPLC

8.4.2.1 Size Exclusion Chromatography

Size exclusion chromatography (SEC) is a chromatographic method in which particles are separated based on their size, or in more technical terms, their hydrodynamic volume. SEC has the potential to separate polymer from its interferential substances before detection. It has proved effective in measuring polymer concentrations in waters containing organic matter, such as runoff waters in oil production fields (Beazley, 1985; Hunt et al., 1988), effluents from coal-washing sites (Leung et al., 1987) and polymer residuals in an alum sludge (Keenan et al., 1998).

8.4.2.2 High Performance Liquid Chromatography

A direct analyses of the polymer residual (artificial polymer solution) was undertaken in the laboratory, based on the mechanism of SEC using HPLC (Keenan et al., 1998). The process uses a chromatographic method in which particles are separated, based on their hydrodynamic volume. Based on the development reported by Keenan et al., (1998), this study employed a high-pressure size exclusion chromatograph (HPSEC), which consists of a Waters 1515 isocratic pump, a Waters 2487 UV dual λ detector operated at 210 nm, and a Rheodyne rotary injection valve equipped with a 20 μl sample loop. A PL Aquagel-OH 40 (300 × 7.5 mm) column was also used. The column was eluted at a flow rate of 1.3 ml/min with a solution of 0.1 M KH_2PO_4 (passed through a 0.45 μm filter) as the mobile phase. The wavelength of the UV detector was set at 210 nm. The injection volume was 20 μl. A photograph of the HPSEC used in this study is shown in Fig. 8.9.

Figure 8.9. Photograph of the Waters HPLC used in this study.

8.4.2.3 Absorbance Response of Polymer in HPLC

Magnafloc LT 25 is an anionic organic polymer, which is widely used in water-purification sludge conditioning. This polymer has molecular weight of 10^{-15}×106 (CIBA Speciality Chemicals Ltd. UK). During the investigation, the polymer was collected from the Ballymore-Eustace water treatment plant where it has been used for years in the sludge-conditioning unit. Under the HPLC operating conditions described above, the retention time for Magnafloc LT 25 solution was approximately 3.67 min. Figure 8.10 illustrates a typical absorbance response of Magnafloc LT 25 in HPLC used in this study.

8.4.2.4 Calibration of Magnafloc LT 25

The calibration of the Magnafloc LT 25 was conducted in the HPLC system. The concentration was calibrated and quantified, based on its peak area.

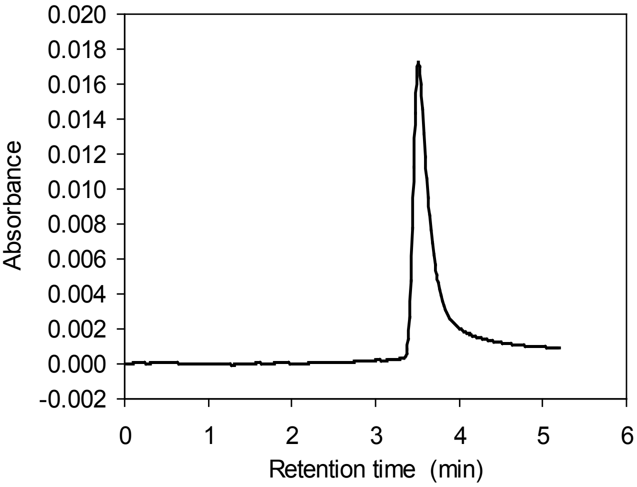


Figure 8.10. Typical response of Magnafloc LT25 on high pressure size exclusion chromatography used in this study.

8.4.3 Monitoring of polymer (Magnafloc LT 25) residual/release from alum sludge reuse

8.4.3.1 Determination of Magnafloc LT-25 in the Effluents by Batch Study and Continuous Flow Column Test

The possible polymer (Magnafloc LT 25) release from dewatered alum sludge was investigated using batch studies. The experiment was conducted by equilibrating 10 g of prepared sludge samples (Section 4.1.1) with 100 ml tap water in a 150 ml plastic bottle which was placed on a Stuart Orbital Shaker (SSL 1, Bibby Sterilin Ltd.) and agitated at 200 rpm for 0.5 hrs, 3 hrs, 10 hrs, 17 hrs, 24 hrs, and 31 hrs. After agitation, the solution was filtered by a 0.45 μm membrane and the filtrate was subjected for polymer residual measurement via HPLC under the conditions described in Section 8.4.2.2.

In order to further investigate the extent of polymer release in a continuous flow environment, a test was conducted in an alum sludge-packed column with tap water flowing through to eliminate the effects of other substances from the influent. The column was as described in Section 5.1 and shown in Fig 8.11. The total mass of the sludge was 7.8 kg. The flow rate of influent (i.e. tap water) was set as 48 ml/min. Effluent samples from the column were collected periodically and analysed for polymer LT 25 using HPLC, following the procedure as described in Section 8.4.2.2 above.

Figure 8.11. Continuous flow test system for polymer-release monitoring.

The results of batch tests to investigate polymer release from dewatered alum sludge are shown in Table 8.4. It can be seen from Table 8.4 that no polymer was observed using agitation times of 0.5 and 3 hrs. The concentration of LT25 remains below 0.1 mg/L with an agitation time of 10 hrs, but increases to 0.13 mg/L after 17 hrs agitation, 0.16 mg/L after 24 hrs agitation, and 0.14 mg/L at 31 hrs agitation. Beyond 17 hrs agitation, the concentration of polymer released does not seem to increase with further agitation time, which implies a polymer release from the sludge to solution, but the amount is insignificant.

Table 8.4. Release of Magnafloc LT 25 from alum sludge with agitation time in batch study.

Agitation time (hrs)	Peak area (mV/s)	Polymer LT 25 concentration (mg/L)
0.5	0	0
3.0	0	0
10.0	1784	Below 0.1
17.0	4017	0.13
24.0	4551	0.16
31.0	4194	0.14

The results of polymer monitoring from the effluent of the sludge column operated by tap water indicated that considerable UV absorbance at the retention time of 3.4 min to 3.8 min was found in the HPLC response curve. After 2 days' continuous flow operation, the concentration of Magnafloc LT 25 in the effluent is 0.83 mg/L while after 3 days' continuous flow operation, the concentration of LT 25 was 0.17 mg/L. Polymer was not detected on the fourth and fifth days.

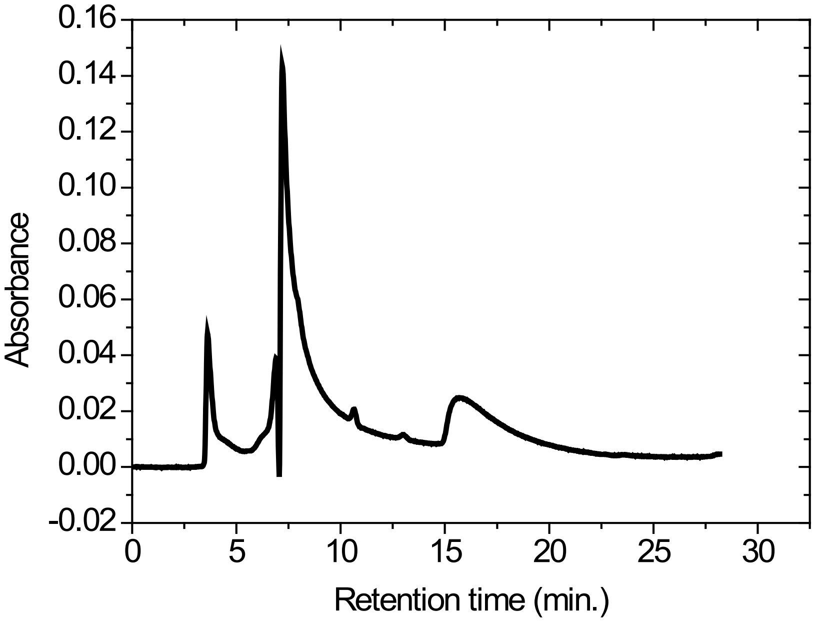


Figure 8.12. High-pressure size exclusion chromatography curve of the effluent of Stage 1 in the 4-stage alum sludge-based CW system.

Figure 8.12 shows the HPLC response of the effluent collected from Stage 1 of the CW. From the figure, it can observed that certain compounds in the wastewater interfere with the LT 25 since the extensive peak at about 3.67 min. retention time makes it impossible to detect the LT 25. The compounds that interfere with the polymer monitoring might be polysaccharides, which are also macromolecular compounds. To further investigate the interference of the substances contained in farmyard wastewater, the supernatant of the farmyard wastewater used as the influent (raw wastewater) in the CW system was subjected to HPLC monitoring. The result is shown in Fig. 8.13. It can be seen that the farmyard wastewater has some compounds with similar molecular size to LT 25 and these compounds can interfere with the LT 25 monitoring in HPLC.

Figures 8.12 and 8.13 both indicate that the LT 25 in the effluent, if any, cannot be separated from interfering substances in the farmyard wastewater under these conditions of chromatography. The HPLC operational conditions such as flow rate, pH of the mobile phase, ion strength of mobile phase, organic modifier used (such as acetonitrile), surface additives (such as *n*-pentanesulfonic acid) were varied. So far, all efforts to achieve a reasonable HPLC response for LT 25 have failed. Therefore, it appears that under current operation conditions, the HPLC cannot reliably detect the polymer LT 25 in the wastewater samples.

8.4.4 Summary

Based on the mechanism of size exclusion chromatography using HPLC, polymer (Magnafloc LT25) residual/release from used alum sludge was monitored. Established methodology of the HPLC system demonstrated a clear response of polymer Magnafloc LT 25 using batch tests and a tap-water operated single alum sludge column. From the batch study, it is reasonable to conclude that the release of polymer from the sludge into solutions increased with the increase of agitation time. During the single sludge column operation, it seems that the polymer release occurs most likely in the first few days of operation. No polymer can be detected after the fourth day of operation and the amount of polymer release is small.

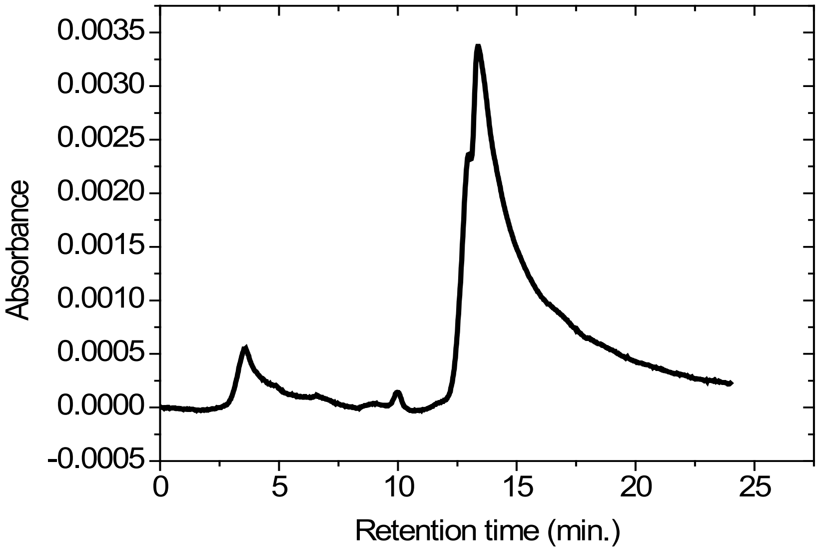


Figure 8.13. High-pressure size exclusion chromatography curve of the supernatant of the farmyard wastewater.

9 Discussion

In view of the wastewater treatment in rural areas and isolated industrial estates, CW is one of the promising technologies to provide cost-effective wastewater treatment alongside sustainable development. In particular, increasing demands for decentralised systems for sewage treatment in rural areas and industrial settings offer great opportunity for the wide application of CW. At the CW research group at UCD, one of the central research goals is the development of a new generation of CW systems that are simple and sustainable in construction, but more efficient than conventional CW systems. The CW research group is the pioneer of the novel alum sludge-based CW system for wastewater treatment which has the capacity for enhanced removal of P and organic matter, particularly from high-strength wastewaters. A special novelty of the newly developed system is the use of a hitherto landfill designated by-product 'dewatered alum sludge' as the substrate in the CW as opposed to the traditional media of gravel, sand and local soils.

9.1 Novelty

It is well known that the substrate in CWs plays a key role in wastewater treatment in these systems. Typically, local soils, sand and gravel have been used as substrates in conventional CW systems. However, while such materials are good carriers of biofilm development, they have poor P removal ability. Consequently, the discovery of an alternative substrate with considerable capacity for P adsorption in CWs is an effective research topic in CW development. Several materials have been tested as a possible substrate for CWs, particularly for enhancing P removal. However, the scale and economics of their use have not been justified and attempts to scale up such preliminary investigation to field scale have been lacking. However, the use of alum sludge as a substrate is justifiable both economically and environmentally. Alum sludge is an inevitable by-product largely produced in water treatment plants worldwide and often designated for landfill disposal. It is locally and easily available free of charge.

9.2 Benefits

There are a number of benefits associated with the use of alum sludge in CWs. They include:

1. Abundant supply of dewatered alum sludge which implies that sufficient quantities of alum sludge for developing alum sludge-based CWs are guaranteed;
2. Reduced capital investment of construction and, at the same time, a drastic reduction in the frequency and cost of media replacement;
3. Good P-adsorption capacity which will conveniently outlast most other CW substrates;
4. Good carrier/material for biofilm attachment and therefore the ability to support biofilm growth and development;
5. Possibility of treating high-strength wastewater because the alum sludge serves to greatly enhance P removal while the tidal flow operation strategy enhances oxygen transfer, leading to greater microbial degradation;
6. Suitable growth medium for wetland plants;
7. Considerable saving in landfill cost and the need for landfill capacity.

9.3 Constraints

Even though the alum sludge-based CW system has demonstrated obvious advantages over conventional CW, there are constraints that should be carefully considered for the large-scale applications. These include:

1. Logistics of the use of the alum sludge at field scale vis-à-vis the application mode of the alum sludge and the typical P-asorption capacity of the alum sludge in the field. As an unconventional substrate material for CW systems, the application mode of the alum sludge (i.e. either freshly dewatered alum sludge; aged dewatered alum sludge; dried alum sludge; granulated alum sludge) should be considered carefully as this will influence the transport and handling logistics. Furthermore, adequate attention should be paid to the P-loading capacity of the alum sludge used in a full-scale CW as this would differ from computations based on laboratory work.
2. The lifespan of the alum sludge regarding its saturation with P (although it has been preliminarily estimated as 4 to 17 years [Zhao et al., 2009]). This can be used as a guide, but it is yet to be fully verified on a field scale.
3. The CW will likely require regular Al monitoring as the effects of Al level beyond the recommended safe limit could be lethal. This may imply additional cost for the CW operation.
4. Clogging is an inevitable long-term operational drawback of CW systems, and it is important to be aware of this as a possible operational challenge in the long term.
5. Eventual disposal of the P-saturated alum sludge, which needs to be planned for. However, as noted above, it is possible to use the saturated sludge as a soil modifier (Babatunde and Zhao, 2009) or for P recovery (Zhao and Zhao, 2009).

10 Conclusions

The sludge derived from the drinking-water treatment process is an inescapable by-product and has been historically viewed as a 'waste' for landfill. Alum sludge, an aluminium salt-coagulated residual, has exhibited excellent phosphorus immobilisation abilities and has the potential for reuse to enhance P removal during wastewater treatment. Based on this concept, this project examined the characteristics of dewatered alum sludge and developed a novel alum sludge-based CW system for high strength wastewater treatment. The main conclusions that can be drawn are listed below.

Dewatered alum sludge can be a good material for use in CWs as the main substrate to improve the treatment performance for both organics and nutrients, especially P because:

1 It is a good low-cost adsorbent for P immobilisation;

2 It can be a good support for biofilm development, thus providing a good environment for degradation of organics in the wetland system;

3 It is a suitable growth medium for common reed, *Phragmites australis,* the most popular wetland plant, growth.

The alum sludge-based CW system is capable of treating high-strength wastewater with comparable high efficiency compared with conventional CW systems, such as gravel-based wetland systems. A long-term run of a single model alum sludge-based CW achieved average removal efficiencies of 73.3±15.9% for COD, 82.9±12.3% for BOD_5, 86.4±6.0% for RP, 88.6±7.2% for SRP and 77.6±17.5% for SS. Results also revealed that the 'P-adsorption proportion' by the sludge in the CW is 42% of overall P removal. Therefore, the lifetime of the alum sludge in the systems is considerably longer than that determined from a batch isotherm test.

A 4-stage CW system using alum sludge as the main substrate was conceptualised, designed and operated to further develop the system. It was shown that the system can enhance the concurrent removal of P and organic matter from wastewater. The system achieved very high removal efficiencies for BOD_5 (90.6%), COD (71.8%), RP (80%) and SRP (89%). In all cases, the first stage of the system contributed most significantly to the overall removal of the pollutants, particularly for soluble reactive P which had 89% of the overall removal achieved in the first stage compared to 53% and 39% removal achieved for BOD_5 and COD respectively, in the first stage.

The field study validated the laboratory results. The field-scale system treating real wastewater emanating from an animal farm exhibited good performance with up to 70% removal of COD, 85% NH_4-N and over 90% phosphorus achieved.

Environmental and health concerns regarding alum sludge reuse were also investigated in the project. Experiments have revealed that the Al in the sludge could be released in the effluent from the CW system, especially in the initial operation period. However, the concentration of Al in the leachates in most cases is below the limits of 0.2 mg/L. Such release does not appear to pose an imminent ecological or environmental risk. Accordingly, concerns about Al release should not restrict its use. However, periodical monitoring is recommended. Similarly, organic polymer was detected in the initial operation period of the simulated alum sludge column, indicating its release. However, such release cannot be traced after 4 days' operation. Like other CW systems, clogging of the substrate might be a problem and this should be considered for the large-scale application of the alum sludge-based CW system. Phosphorus can be recovered from the saturated alum sludge in the CW system. A recovery methodology was established in the study. In addition, the used alum sludge could offer an alternative option as fertiliser for P-deficient soils. Due to the nature of the geographic distribution of residents of Ireland, CW has huge potential for application of small wastewater treatment systems and the newly developed CW can be readily deployed in such situations.

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Acronyms and Annotations

BOD_5	5-day biochemical oxygen demand. A primary key water quality indicator that is used as a measure of the quantity of oxygen used by microorganisms in the oxidation of organic matter
COD	Chemical oxygen demand. A primary key water quality indicator that is used as a measure of the oxygen requirement of a sample that is susceptible to oxidation by a strong chemical oxidant
CW	Constructed Wetlands. A green wastewater treatment technique that mimics the purification processes in natural wetlands
d_{10}	Diameter of the largest particle in the smallest 10% (by mass) of particles of the sample
d_{60}	Diameter of the largest particle in the smallest 60% (by mass) of particles in the sample
EC	Electrical conductivity. An estimate of the total dissolved salts or the total amount of dissolved ions in water
Eh	Oxidation-reduction potential. The measure of a chemical species to acquire or donate electrons
EDS	Energy-dispersive X-ray spectroscopy. An analytical technique used for the elemental analysis or chemical analysis of samples
FTIR	Fourier transform infrared spectroscopy. A technique used to obtain an infrared spectrum of absorption, emission, photoconductivity or Raman scattering of a solid, liquid or gas.
HPLC	High performance liquid chromatography or high pressure liquid chromatography. A chromatographic technique that can separate a mixture of compounds, and is used in biochemistry and analytical chemistry to identify, quantify and purify the individual components of the mixture
HPSEC	High pressure size exclusion chromatography. A technique used to evaluate the molecular sizes of humic substances from different sources
ICP-MS	Inductively coupled plasma mass spectrometry. A technique used for the determination of trace elements in solution
ICP-OES	Inductively coupled plasma optical emission spectrometry. A fast multi-element technique with a dynamic linear range and moderately low detection limits
LOI	Loss of ignition. A test used to give a crude measure of the organic content of a sample
NH_4-N	Ammonium nitrogen. A primary indicator of water quality
NTU	Nephelometric turbidity units. A unit of measurement of turbidity
PSD	Particle size distribution. The percentage of each fraction into which a sample is classified, with respect to particle size, by number or weight
RP	Reactive phosphorus. A measure of orthophosphate that is determined directly without any pre-treatment
SCOD	Soluble chemical oxygen demand
SEC	Size exclusion chromatography
SRP	Soluble reactive phosphorus. A measure of orthophosphate, the filterable (soluble, inorganic) fraction of phosphorus, the form directly taken up by plant cells
SS	Suspended solids. A water quality measurement which refers to small solid particles which remain in suspension in water

SSA	Specific surface area. A material property of solids which measures the total surface area per unit of mass, solid or bulk volume, or cross-sectional area
TOC	Total organic carbon. The amount of carbon bound in an organic compound and is often used as a non-specific indicator of water quality
UC	Uniformity coefficient. This is defined as a ratio: the size at which 60% (by mass) of a sample passes through a sieve (in other words, 60% of the material is finer than a given size) divided by the size at which 10% of the sample (by mass) passes through a sieve (10% is finer than a given size)

Appendix: Publication List

I. Peer-Reviewed Journals

1. Babatunde, A.O., Zhao, Y.Q. 'Phosphorus removal in laboratory-scale unvegetated vertical subsurface flow constructed wetland systems using alum sludge as main substrate', *Water Science and Technology*, 60(2), 483–489, 2009 (ISSN: 0273–1223)
2. Babatunde, A.O., Zhao, Y.Q. 'Forms, patterns and extractability of phosphorus retained in alum sludge used as substrate in laboratory-scale constructed wetland systems', *Chemical Engineering Journal,* 152(1), 8–13, 2009 (ISSN: 1385-8947)
3. Babatunde, A.O., Zhao. Y.Q., Burke, A.M., Morris, M.A., Hanrahan, J.P. '**C**haracterization of aluminium-based water treatment residual for potential phosphorus removal in engineered wetlands', *Environmental Pollution,* 157(10), 2830–2836, 2009 (ISSN**:**0269-7491)
4. Zhao, X.H., Zhao, Y.Q. 'Decolouration of H_2SO_4 leachate from phosphorus-saturated alum sludge using H_2O_2 and advanced oxidation processes in phosphorus recovery strategy', *Journal of Environmental Science and Health, Part A: Toxic/Hazardous Substances & Environmental Engineering*, 44(14), 1557–1564, 2009 (ISSN: 1093–4529)
5. Zhao, Y.Q, Babatunde, A.O., Zhao, X.H., Li, W.C. 'Development of alum sludge-based constructed wetland: An innovative and cost-effective system for wastewater treatment', *Journal of Environmental Science and Health, Part A: Toxic/Hazardous Substances & Environmental Engineering*, 44(8), 827–832, **2009** (ISSN: 1093–4529)
6. Zhao, Y.Q., Babatunde, A.O., Zhao, X.H. 'An innovative solution for managing waterworks sludge: Developing an alum sludge-based multi-stage constructed wetland system for wastewater treatment', *Journal of ASTM International*, 6(6), 2009 (ISSN: 1546–962X)
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8. Zhao, X.H., Zhao, Y.Q. 'Investigation of phosphorus desorption from P-saturated alum sludge used as a substrate in constructed wetland', *Separation and Purification Technology,* 66(1), 71–75, 2009 (ISSN: 1383–5866)
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10. Babatunde, A.O., Zhao, Y.Q., O'Neill, M., O'Sullivan, B. 'Constructed wetlands for environmental pollution control: A review of developments, research and practice in Ireland', *Environment International*, 34(1), 116–126, 2008 (ISSN: 0160–4120)
11. Babatunde, A.O., Zhao, Y.Q. 'Constructive approaches towards water treatment works sludge management: An international review of beneficial re-uses', *Critical Reviews in Environmental Science and Technology*, 37(2), 129–164, 2007 (ISSN: 1064-3389)
12. Babatunde, A.O, Zhao, Y.Q., Yang, Y. Kearney, P. 'From 'fills' to filter: Insights into the reuse of dewatered alum sludge as a filter media in a constructed wetland', *Journal of Residuals Science & Technology*, 4(3), 147–152, 2007 (ISSN: 1544-8053)
13. Babatunde, A.O., Zhao, Y.Q. 'A novel alum-sludge based constructed wetland system to reduce pollution effects of agricultural runoff: first results', *International Journal of Water*, 3(3), 207–213, 2007 (ISSN: 1465–6620)

II. Conference Papers

14. Zhao, Y.Q, Babatunde, A.O., Zhao, X.H., Kumar, J.L.G., Hu, Y.S. ‘Alum sludge-based constructed wetland: Novelty, benefits and constraints’, In: Proceedings of *Sustainable Water Infrastructure for Cities and Villages of the Future (SWIF 2009)*, 6–9 November 2009, Beijing, China. 9 pages in CD-Rom.

15. Kumar, J.L.G., Zhao, Y.Q., Zhao, X.H., Babatunde, A.O. ‘Determination of first-order rate constant for phosphorus removal in a novel model-reed bed treatment system’, In: Proceedings of *2009 Beijing International Environmental Technology Conference*, 16–18 October 2009, Beijing, China. 9 pages in CD-Rom.

16. Zhao, Y.Q., Babatunde, A.O., Zhao, X.H. ‘Overview of developing new generation of engineered wetlands for wastewater treatment: First experience from Ireland’, In: Proceedings of the *2nd Taiwan-Mainland China Constructed Wetland Conference*, 18–21 May, 2009, Kaohsiung, Taiwan, pp. 223–232.

17. Zhao, X.H., Zhao, Y.Q., ‘Phosphorus extraction from saturated alum sludge used as a low-cost P-adsorbent’, In: Proceedings of *International Conference on Nutrient Recovery from Wastewater Streams*, 10–13 May, 2009, Vancouver, British Columbia, Canada, 13 pages in CD-Rom (ISBN: 1843392321), IWA Publishing, London (edited by Ken Ashley, Don Mavinic and Fred Koch).

18. Li, W.C., Zhao, Y.Q. ‘An Innovative use of dewatered alum sludge as main substrate in a multi-stage reed bed treatment system’, In: Proceedings of *IWA-ASI Workshop on Sustainability of Water Environment and Water Resources and Perspectives of Energy and Resources Saving and Recovery in Wastewater Treatment*, HKUST, Hong Kong, China, 22–27 June 2008, pp. 95–102.

19. Zhao. Y.Q, Babatunde, A.O., Zhao, X.H., Li, W.C. ‘Alum sludge-based constructed wetland: An innovative and cost-effective system for wastewater treatment’, In: Proceedings of *11th International Conference on Wetland System for Water Pollution Control*, 1–7 November 2008, Indore, India, Vol. II, pp. 621–626.

20. Babatunde, A.O., Zhao. Y.Q. ‘Testing and evaluation of a tidal flow alum sludge based constructed wetland system’, In: Proceedings of *11th International Conference on Wetland System for Water Pollution Control*, 1–7 November **2008**, Indore, India, Vol. I, pp. 192–196.

21. Babatunde, A.O., Zhao, Y.Q. ‘Optimizing phosphorus removal in vertical subsurface flow constructed wetland systems using a drinking-water treatment residual as main substrate’, In: Abstract book of *6th IWA World Water Congress*, 7–12 September 2008, Vienna, Austria, pp. 176–177 (8 pages in CD-Rom).

III. Magazine Articles

22. Babatunde, AO; Zhao, YQ; (2009) New low-cost wastewater treatment system pioneered in Ireland; In: *ICE-Civil Engineering*, Vol. 162(4), pp. 148–148. London, UK

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An Ghníomhaireacht um Chaomhnú Comhshaoil

Is í an Gníomhaireacht um Chaomhnú Comhshaoil (EPA) comhlachta reachtúil a chosnaíonn an comhshaol do mhuintir na tíre go léir. Rialaímid agus déanaimid maoirsiú ar ghníomhaíochtaí a d'fhéadfadh truailliú a chruthú murach sin. Cinntímid go bhfuil eolas cruinn ann ar threochtaí comhshaoil ionas go nglactar aon chéim is gá. Is iad na príomh-nithe a bhfuilimid gníomhach leo ná comhshaol na hÉireann a chosaint agus cinntiú go bhfuil forbairt inbhuanaithe.

Is comhlacht poiblí neamhspleách í an Ghníomhaireacht um Chaomhnú Comhshaoil (EPA) a bunaíodh i mí Iúil 1993 faoin Acht fán nGníomhaireacht um Chaomhnú Comhshaoil 1992. Ó thaobh an Rialtais, is í an Roinn Comhshaoil agus Rialtais Áitiúil a dhéanann urraíocht uirthi.

ÁR bhFREAGRACHTAÍ

CEADÚNÚ

Bíonn ceadúnais á n-eisiúint againn i gcomhair na nithe seo a leanas chun a chinntiú nach mbíonn astuithe uathu ag cur sláinte an phobail ná an comhshaol i mbaol:

- áiseanna dramhaíola (m.sh., líonadh talún, loisceoirí, stáisiúin aistrithe dramhaíola);
- gníomhaíochtaí tionsclaíocha ar scála mór (m.sh., déantúsaíocht cógaisíochta, déantúsaíocht stroighne, stáisiúin chumhachta);
- diantalmhaíocht;
- úsáid faoi shrian agus scaoileadh smachtaithe Orgánach Géinathraithe (GMO);
- mór-áiseanna stórais peitreail.
- Scardadh dramhuisce

FEIDHMIÚ COMHSHAOIL NÁISIÚNTA

- Stiúradh os cionn 2,000 iniúchadh agus cigireacht de áiseanna a fuair ceadúnas ón nGníomhaireacht gach bliain.
- Maoirsiú freagrachtaí cosanta comhshaoil údarás áitiúla thar sé earnáil - aer, fuaim, dramhaíl, dramhuisce agus caighdeán uisce.
- Obair le húdaráis áitiúla agus leis na Gardaí chun stop a chur le gníomhaíocht mhídhleathach dramhaíola trí comhordú a dhéanamh ar líonra forfheidhmithe náisiúnta, díriú isteach ar chiontóirí, stiúradh fiosrúcháin agus maoirsiú leigheas na bhfadhbanna.
- An dlí a chur orthu siúd a bhriseann dlí comhshaoil agus a dhéanann dochar don chomhshaol mar thoradh ar a ngníomhaíochtaí.

MONATÓIREACHT, ANAILÍS AGUS TUAIRISCIÚ AR AN GCOMHSHAOL

- Monatóireacht ar chaighdeán aeir agus caighdeáin aibhneacha, locha, uiscí taoide agus uiscí talaimh; leibhéil agus sruth aibhneacha a thomhas.
- Tuairisciú neamhspleách chun cabhrú le rialtais náisiúnta agus áitiúla cinntí a dhéanamh.

RIALÚ ASTUITHE GÁIS CEAPTHA TEASA NA HÉIREANN

- Cainníochtú astuithe gáis ceaptha teasa na hÉireann i gcomhthéacs ár dtiomantas Kyoto.
- Cur i bhfeidhm na Treorach um Thrádáil Astuithe, a bhfuil baint aige le hos cionn 100 cuideachta atá ina mór-ghineadóirí dé-ocsaíd charbóin in Éirinn.

TAIGHDE AGUS FORBAIRT COMHSHAOIL

- Taighde ar shaincheisteanna comhshaoil a chomhordú (cosúil le caighdéan aeir agus uisce, athrú aeráide, bithéagsúlacht, teicneolaíochtaí comhshaoil).

MEASÚNÚ STRAITÉISEACH COMHSHAOIL

- Ag déanamh measúnú ar thionchar phleananna agus chláracha ar chomhshaol na hÉireann (cosúil le pleananna bainistíochta dramhaíola agus forbartha).

PLEANÁIL, OIDEACHAS AGUS TREOIR CHOMHSHAOIL

- Treoir a thabhairt don phobal agus do thionscal ar cheisteanna comhshaoil éagsúla (m.sh., iarratais ar cheadúnais, seachaint dramhaíola agus rialacháin chomhshaoil).
- Eolas níos fearr ar an gcomhshaol a scaipeadh (trí cláracha teilifíse comhshaoil agus pacáistí acmhainne do bhunscoileanna agus do mheánscoileanna).

BAINISTÍOCHT DRAMHAÍOLA FHORGHNÍOMHACH

- Cur chun cinn seachaint agus laghdú dramhaíola trí chomhordú An Chláir Náisiúnta um Chosc Dramhaíola, lena n-áirítear cur i bhfeidhm na dTionscnamh Freagrachta Táirgeoirí.
- Cur i bhfeidhm Rialachán ar nós na treoracha maidir le Trealamh Leictreach agus Leictreonach Caite agus le Srianadh Substaintí Guaiseacha agus substaintí a dhéanann ídiú ar an gcrios ózóin.
- Plean Náisiúnta Bainistíochta um Dramhaíl Ghuaiseach a fhorbairt chun dramhaíl ghuaiseach a sheachaint agus a bhainistiú.

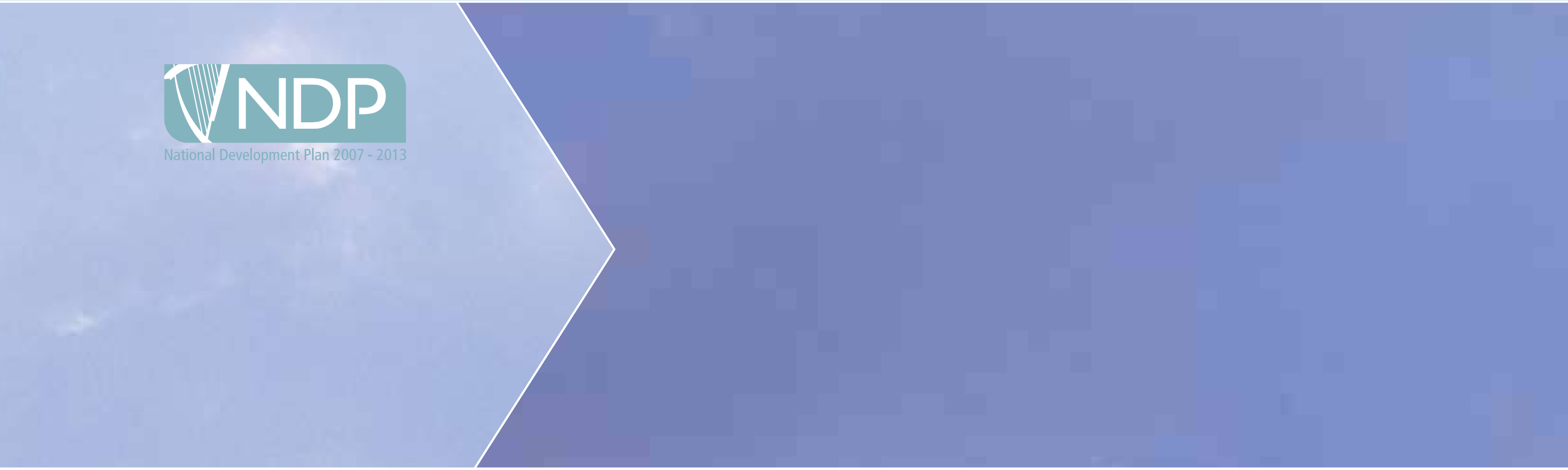
STRUCHTÚR NA GNÍOMHAIREACHTA

Bunaíodh an Ghníomhaireacht i 1993 chun comhshaol na hÉireann a chosaint. Tá an eagraíocht á bhainistiú ag Bord lánaimseartha, ar a bhfuil Príomhstiúrthóir agus ceithre Stiúrthóir.

Tá obair na Gníomhaireachta ar siúl trí ceithre Oifig:

- An Oifig Aeráide, Ceadúnaithe agus Úsáide Acmhainní
- An Oifig um Fhorfheidhmiúchán Comhshaoil
- An Oifig um Measúnacht Comhshaoil
- An Oifig Cumarsáide agus Seirbhísí Corparáide

Tá Coiste Comhairleach ag an nGníomhaireacht le cabhrú léi. Tá dáréag ball air agus tagann siad le chéile cúpla uair in aghaidh na bliana le plé a dhéanamh ar cheisteanna ar ábhar imní iad agus le comhairle a thabhairt don Bhord.



NDP
National Development Plan 2007 - 2013

Science, Technology, Research and Innovation for the Environment (STRIVE) 2007-2013

The Science, Technology, Research and Innovation for the Environment (STRIVE) programme covers the period 2007 to 2013.

The programme comprises three key measures: Sustainable Development, Cleaner Production and Environmental Technologies, and A Healthy Environment; together with two supporting measures: EPA Environmental Research Centre (ERC) and Capacity & Capability Building. The seven principal thematic areas for the programme are Climate Change; Waste, Resource Management and Chemicals; Water Quality and the Aquatic Environment; Air Quality, Atmospheric Deposition and Noise; Impacts on Biodiversity; Soils and Land-use; and Socio-economic Considerations. In addition, other emerging issues will be addressed as the need arises.

The funding for the programme (approximately €100 million) comes from the Environmental Research Sub-Programme of the National Development Plan (NDP), the Inter-Departmental Committee for the Strategy for Science, Technology and Innovation (IDC-SSTI); and EPA core funding and co-funding by economic sectors.

The EPA has a statutory role to co-ordinate environmental research in Ireland and is organising and administering the STRIVE programme on behalf of the Department of the Environment, Heritage and Local Government.

epa
Environmental Protection Agency
An Ghníomhaireacht um Chaomhnú Comhshaoil

ENVIRONMENTAL PROTECTION AGENCY
PO Box 3000, Johnstown Castle Estate, Co. Wexford, Ireland
t 053 916 0600 f 053 916 0699
LoCall 1890 33 55 99
e info@epa.ie w http://www.epa.ie

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Strategy for Science, Technology and Innovation

Comhshaol, Oidhreacht agus Rialtas Áitiúil
Environment, Heritage and Local Government