

## Removing phosphate from a municipal wastewater treatment plant in the United Kingdom using a waste-to-resource model

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### Abstract

The dispersal of phosphorus from sewage, industrial waste, detergents, and urban and agricultural runoff has played a major part in the eutrophication of many freshwater and marine ecosystems in Europe. As a consequence, phosphate discharge consents on municipal and industrial effluent are being tightened to  $\leq 2.0$  mg/L. Many agricultural activities, industrial processes and water treatment companies therefore face additional requirements to reduce soluble phosphate in discharge waters. A new green filtration technology, which repurposes alumina refinery residue, offers a passive, flow-through treatment system for phosphorus removal. The technology has been shown elsewhere to reduce or eliminate the need for chemical dosing, allow for variations in hydraulic flow and nutrient loading, minimise sludge production, and provide an effective and straightforward treatment system when applied immediately prior to final effluent discharge.

A four-month trial to evaluate the potential use of this waste-to-resource technology to enhance the removal of phosphate from municipal wastewater in line with these tighter standards was conducted in the United Kingdom. The primary objective was to determine if the technology could reliably maintain phosphate concentrations to extremely low consent limits. The trial was conducted at Yorkshire Water's Kirk Smeaton wastewater treatment plant in North Yorkshire, and comprised three separate filter configurations in order to provide comparison data on different design scenarios, hydraulic residence times, removal efficiencies and projected filter life-spans.

Filter Pairing A consisted of two columns operating in series with a hydraulic residence time of 12 hours. Influent phosphate concentrations averaged 10.3 mg/L for the duration of the trial and effluent concentrations averaged 2.6 mg/L, a removal efficiency across both columns of 74%. In addition, biological oxygen demand was reduced from 6.8 mg/L to 4.3 mg/L, and ammonia-nitrogen was reduced from 1.1 mg/L to 0.86 mg/L. Filter Pairing B again consisted of a two-stage filter system operating in series but with a shorter hydraulic residence time of two to three hours. Influent phosphate concentrations averaged 9.6 mg/L and effluent concentrations averaged 1.8 mg/L, a removal efficiency across both columns of 81%. Filter Pairing C, consisted of a three-stage filter system operated in series with a hydraulic residence time of 12 hours, but in contrast with Filter Pairing A each filter operated with a hydraulic residence time of four hours. Influent phosphate concentrations averaged 9.6 mg/L and effluent concentrations averaged 0.37 mg/L, a phosphate removal efficiency across both columns of 96%.

Results of this study indicate that Filter Pairings B and C achieved phosphate removal efficiencies which conform to the tighter regimes being imposed throughout the UK and Europe.

**Keywords:** Municipal wastewater, alumina refinery residue, phosphate, filtration, sewage treatment, waste-to-resource

### Introduction

The widespread dispersal of phosphorus from sewage effluent, industrial wastewater, detergents, and run-off from urban and agricultural land has played a major part in the eutrophication of many freshwater and marine ecosystems throughout Europe (European Environment Agency, 2000; Withers *et al.*, 2014)<sup>[17, 33]</sup>, and was the subject of the Urban Waste Water Treatment Directive 91/271.EEC as early as 1991 (Department of Environment, Food and Rural Affairs, 2012). The relationship between phosphorus and eutrophication has been known for many years (e.g., Bowles & Quennell, 1971; Cartwright *et al.*, 1993, Lapointe *et al.*, 2015)<sup>[4, 7, 26]</sup>, and the role of sewage effluent, above other point and diffuse sources of phosphorus in water pollution, has been well documented (Jarvie *et al.*, 2006)<sup>[25]</sup>. [For a more detailed description of the trophic states of eutrophication and their relation to environmental degradation as delineated by the Organisation for Economic Co-operation and Development, see Diston *et al.*, 2007.]<sup>[11]</sup> In order to help tackle eutrophication in inland and coastal waters, more stringent measures are being applied to, for

example, agricultural pollution (Environmental Data Services, 2006a)<sup>[13]</sup> and detergents (Commission of the European Communities, 2007)<sup>[8]</sup>, and stricter discharge consents are being imposed by environmental regulators on total phosphate (TP) concentrations in effluent from wastewater treatment plants (WTPs). Phosphate consents (or licenses to discharge effluent into receiving creeks, lakes or rivers) have dropped (or will drop) in the UK from typically between 5.0-10.0 mg/L to  $\leq 2.0$  mg/L under Directive 91/271.EEC. These limits may be further tightened to  $\leq 1.0$  mg/L in more eco-sensitive areas, as recommended two decades ago by English Nature and other agencies (Diston *et al.*, 2007; Environmental Data Services, 1997; Water Briefing, 2014)<sup>[11, 12, 31]</sup> and limits as low as  $\leq 0.5$  mg/L have been considered.

The need for stricter TP consents at the UK's 9,000 WTPs (Department of Environment, Food and Rural Affairs, 2012, p. 7) is supported by influential studies on seven lowland river catchments, which concluded that phosphate in sewage effluent presents a greater eutrophication risk to the environment than agricultural run-off (e.g., Jarvie *et al.*, 2006)<sup>[25]</sup>. This study

found that a strong correlation between soluble phosphate emissions and algal growth was observed at 54 monitoring sites in the UK, with sewage effluent being the main source of eutrophication. Boni *et al.* (1991) [2] had earlier shown that municipal wastewater has a high eutrophication effect, even after biological treatment, and that a deterioration of a WTP's phosphate removal efficiency occurs after heavy rainfall. Similarly, inadequate control of aeration that might occur over weekends when operators are not present or during rain events, for example, also has an adverse effect on phosphate removal efficiencies (Brdjanovic *et al.*, 1998) [5], hence the need for strict operating standards and ways to supplement biological treatment.

Research conducted by the Centre for Ecology and Hydrology in the UK has shown that the removal of phosphates from the effluent streams of smaller, rural WTPs could be more important than from larger WTPs, as smaller WTPs are often closer to headwaters and therefore the discharge of phosphate has a more profound impact on downstream nutrient loadings in creeks and rivers and along sensitive tributaries (Bowes *et al.*, 2005) [3]. These and other findings have led to a significant increase in the number of water bodies designated as "sensitive" to algal growth and toxic blooms (e.g., Environmental Data Services, 2006b) [14], with 297 sensitive areas having been declared in England alone in 1994, a number which had been increased to 588 by 2012 (Department of Environment, Food and Rural Affairs, 2012, p. 14) [10]. Added to this challenge for smaller WTPs, whose discharges apparently represent the largest environmental impact, is the fact that smaller sites are often unmanned but also attract the tightest consents due to their eco-sensitive locations near farmlands and at inland headwaters. However, problems with removing phosphate from municipal wastewater remains a challenge, with 16 billion litres of untreated sewage entering the River Lee in the Stratford area of London alone in 2012 (Department of Environment, Food and Rural Affairs, 2012, p. 5) [10].

The move to more tightly control TP concentrations in wastewater and other sources of nutrient contamination have come from the Department of Environment, Food and Rural Affairs (DEFRA), the Water Services Regulation Authority (Ofwat), and the UK Technical Advisory Group (UKTAG) (Environmental Data Services, 2006c) [15], as well as from the aforementioned water Directive of the European Union. For example, UKTAG advised the government in the 1990s that 65% of England's rivers failed phosphate limits, with lakes being more sensitive to contamination; UKTAG predicted that up to 70% of English lakes and up to 25% of Scottish lakes risk failure of approved phosphate limits. In the UK, UKTAG estimated that 50% of phosphate contamination originated with agriculture, 35% from sewage, and 7% from industry (Environmental Services Data, 2006c) [15].

As a consequence of these developments, water companies have been faced with the prospect of implementing additional treatment methods in order to supplement their traditional biological, chemical and/or physical processes. However, finding a viable method for reducing TP can be problematic, particularly when low consents are required and when the WTP is at a remote, unmanned location, which is the case for about 7,000 of the UK's WTPs. The challenge for industry has been to identify treatment options for phosphate removal which can cost-effectively and reliably achieve these higher quality

treatment standards. Notwithstanding the potential costs, the Environment Agency predicted that 500 WTPs would have phosphate removal equipment fitted by 2010 (Environmental Data Services, 2006c) [15], although with lower rates of conversion than other European states it is unclear how many achieved actually this goal in the UK (Diston *et al.*, 2007) [11]. For example, Anglian Water reported that 73 of its 1,077 WTPs have been fitted at a cost of £50 million, Wessex Water reported that 21 of its WTPs have been fitted at a total cost of £15 million with a further 17 to be fitted in the next few years, and Welsh, Southwest, Severn Trent, Northumbrian and Southern Water reported they had fitted phosphate removal equipment to 89 WTPs with a further 113 to follow. Such changes are not without significance. Neil *et al.* (2000) [27] have shown the addition of chemical dosing at the Marlborough WTP, which discharges into the River Kennet, reduced soluble reactive phosphorus by as much as 75%, producing an effluent that was comparable in TP loads to background river levels upstream of the plant.

The main method employed for phosphate removal at WTPs is a combination of physical separation, chemical dosing, and biological nutrient reduction. Depending on the size and configuration of the plant, the process methods employed by the operator, the tightness of consent levels, and whether heavy metal limits are also applied to effluent discharge consents, these standard approaches result in varying phosphate removal efficiencies, and can be summarised by the following:

**Physical Separation:** Virtually every WTP in the world utilises some form of solid-liquid separation. In so doing, phosphate can be reduced in the liquid stream as it is materially bound to the solid fraction (or biomass) of sewage wastewater. However, physical separation alone only results in minimal phosphate reduction in the liquid phase because most phosphate in sewage is present in its soluble form and therefore must be removed from the liquid fraction through additional chemical reaction or biological reduction: physical separation alone cannot reduce TP to  $\leq 2.0$  mg/L.

**Chemical Dosing:** While chemical dosing is generally effective where TP consents are  $> 2.0$  mg/L, chemical dosing (usually with ferric chloride [FeCl<sub>3</sub>]) by direct addition for lower levels of phosphate requires accurate and variable dosing equipment (because effluent quality and quantity change significantly according to diurnal and seasonal flows) with appropriate pH adjustment requiring sophisticated control systems due to the low pH of most chemicals used for this purpose. [The chemical reaction between FeCl<sub>3</sub> and phosphate can be approximated by the following:  $3\text{FeCl}_2 + 2\text{PO}_4^- \rightarrow \text{Fe}_3(\text{PO}_4)_2 + 6\text{Cl}$ .] Moreover, chemical dosing requires: a) strict health and safety handling standards and controls; b) regular operator monitoring and intervention; and c) increased space and bunded storage areas because many of the chemicals used in phosphate treatment are classified as "hazardous" or "dangerous". For example, ferric chloride used widely throughout industry, is corrosive and burns the skin on contact (Brown, 2015) [6]. Treatments using chemical dosing for TP reduction also result in higher sludge volumes, imposing additional handling and treatment costs and potential disposal problems. The threat of overdosing with ferric salts or poly-aluminium chloride (used as an anti-foaming agent) can result in iron, chloride and/or aluminium "carry-over" into the final

discharge, thereby contributing to further environmental risk and breaches of consents.

**Biological Treatment:** Biological treatment requires a steady, biodegradable carbon source in the waste stream, such as brewery or other food waste. Incorporating biological treatment may involve the restructuring of a WTP's treatment process and typically requires chemical dosing as a back-up for fail safety. Moreover, biological treatment, particularly with more sophisticated WTPs such as biological nutrient removal (BNR) processes, can be temperamental to operate and often requires enhanced nutrient balancing (i.e., between dissolved oxygen, nitrates and phosphorus) to avoid the so-called "secondary release" of phosphate back into the liquid waste stream.

Based on these applications, the water industry in the UK has explored simple-to-install, low maintenance, passive treatment alternatives to these conventional approaches for phosphate removal. Such innovation is particularly relevant for smaller, unmanned WTPs where treatment costs, operational logistics and environmental and social sensitivities are often the greatest.

First developed in Australia in 2001, a filtration method using modified alumina refinery residue (MARR) to remove phosphate from wastewater has been applied as both a direct addition and filtration system at small- and medium-sized WTPs (Fergusson, 2010) [21]. For example, the technology has been trialled or implemented in Australia at Brisbane Water, one of the largest water utilities in the country, Richmond Valley Shire Council, Kilcoy Shire Council, Redcliffe Shire Council, the Royal Air Force base at Amberley in Queensland, and Kyogle Shire Council; larger scale, retrofitted applications in the UK have also been investigated (see for example, Figure 1). The primary focus of the technology relates to its ability to reduce TP and heavy metals in discharge effluent, however it has also demonstrated an ability to reduce biological oxygen

demand (BOD), total suspended solids (TSS), pathogens, such as *E.Coli*, and turbidity-colour, and to improve clarity. Modest reductions in nitrogen (specifically, total nitrogen [TN], ammonia [NH<sub>3</sub>-N], and total Kjeldahl nitrogen [TKN]), have also been observed.

When applied using filtration, the technology has been presented as a simple "flow-through" treatment system which can be applied at the back-end of trickling filter, activated sludge (AS) or BNR WTPs (Fergusson, 2004a, 2006a, 2006b; Hogan *et al.*, 2005) [18, 19, 20, 24], and data suggest that it can eliminate the need for sludge treatment and filter cake pressing, thereby reducing the footprint required to treat TP to lower concentrations. The technology can be applied as a stand-alone filtration system or retrofitted to existing assets, and filter cartridges are packed with a porous pelletised media (see for example, Figure 2). The treatment beds or filters can either be configured for horizontal flow, upward vertical flow, or as a gravity-fed trickling-filter to suit each location, and redundant assets (such as lagoons, disused drying beds, or redundant trickling filters) may be retrofitted to accommodate the system. Layout alternatives include flexibility in design (such as submerged horizontal-flow reed beds, intermittently submerged vertical-flow beds, or biological or percolating filters), duty and standby configurations, and parallel or serial operation.

Moreover, it has been argued the technology is an example of the "waste-to-resource" model of urban sustainability because it uses the waste of one industry (in this case, residue from alumina refining) and, after repurposing the waste through industrial modification techniques, reuses the material as the active ingredient in a pelletised filter media to treat the wastewater generated from a variety of municipal and industrial sources. Examples of how this model has been applied across several industrial sectors and applications have been provided elsewhere (Fergusson, 2015) [22].



**Fig 1:** Tertiary settling pond at Wessex Water's Cranborne WTP prior to retrofitting (left); the same settling pond after installation of the technology (right).

The pelletised media used in the technology is a proprietary mixture of MARR and ordinary Portland cement, and is mostly composed of carbonate, hydroxide and hydroxycarbonate minerals. These minerals include hematite (Fe<sub>2</sub>O<sub>3</sub>), boehmite ( $\gamma$ -AlOOH), gibbsite (Al[OH]<sub>3</sub>), sodalite (Na<sub>4</sub>Al<sub>3</sub>Si<sub>3</sub>O<sub>12</sub>Cl), anatase (TiO<sub>2</sub>), aragonite (CaCO<sub>3</sub>), brucite (Mg[OH]<sub>2</sub>), diaspore ( $\beta$ -Al<sub>2</sub>O<sub>3</sub>.H<sub>2</sub>O), ferrihydrite (Fe<sub>5</sub>O<sub>7</sub>[OH].4H<sub>2</sub>O), gypsum (CaSO<sub>4</sub>.2H<sub>2</sub>O), hydrocalumite (Ca<sub>2</sub>Al[OH]<sub>7</sub>. 3H<sub>2</sub>O),

hydrotalcite (Mg<sub>6</sub>Al<sub>2</sub>CO<sub>3</sub>[OH]<sub>16</sub>.4H<sub>2</sub>O) and para-aluminohydrocalcite (CaAl<sub>2</sub>[CO<sub>3</sub>]<sub>2</sub>[OH]<sub>4</sub>.3H<sub>2</sub>O). The chemical properties of these pellets are somewhat unique as they can neutralise between 3.5-7.5 moles of acid/kg, have a high metal binding capacity (up to 1,500 meq of metals/kg), and contain a multi-charged surface for potential phosphate reaction sites. Of significance are the presence of negatively charged hydroxides and oxyhydroxide compounds, as well as the positively charge

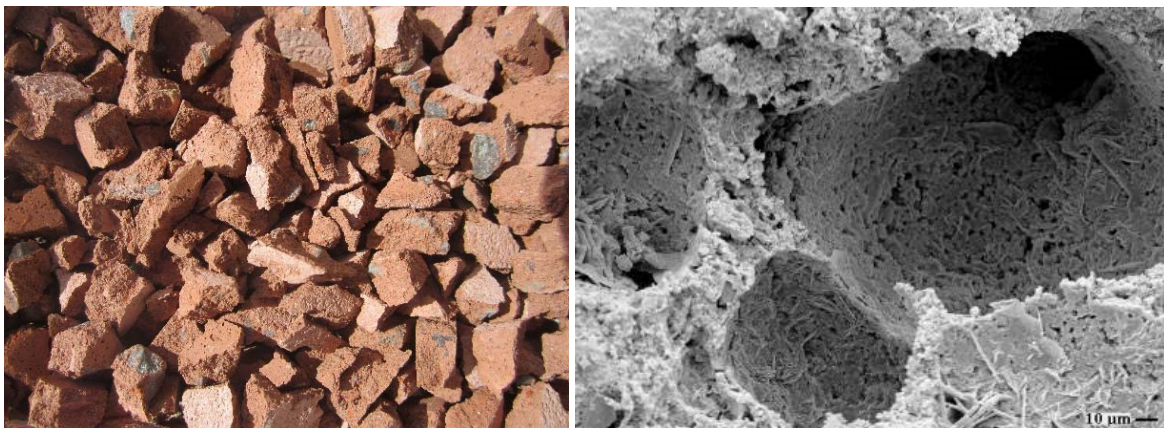
iron-, aluminium-, magnesium- and titanium-based molecules, which adsorb negatively charged phosphate ( $\text{PO}_4^-$ ).

The physical properties of MARR in the pellets are also relevant in that they have a high charge-to-mass ratio and a high surface area-to-mass ratio (about  $300\text{mm}^2/\text{g}$ ), are composed of up to 80% of solid grains ( $10\ \mu\text{m}$  or less) with a pH of about 9.0, are insoluble and non-dispersive in water, display fast settling times when applied as a dry solid with a specific gravity of  $3.2\ \text{g}/\text{cm}^3$ , and produce a stable sediment not a sludge when applied directly to water. Figure 2 shows the generally porous nature of the MARR pellets (i.e., its macroporosity) and the interconnection between pores (i.e., its microporosity) using a scanning electron microscopic (SEM), with large internal and external reactive surfaces adding to the removal of contaminants. Phosphate removal occurs by a variety of ionic and precipitation reactions within the porous pellet matrix; it has also been proposed the pellets may act as an immobilisation substrate for bacteria to promote biological activity.

The effective removal of phosphate to low concentrations using this technology has been achieved with no requirement to add flocculants or adjust influent pH. In addition to TP treatment, the technology also may offer a solution for some of the

problems associated with conventional chemical dosing, producing an effluent reduced in BOD, ammonia, suspended solids, odour and colour, and binds excess iron released in the effluent, thereby preventing iron carry-over. The immobilised pellet bed allows for variations in hydraulic flow and TP concentrations, and upstream chemical dosing is typically not required. In addition to phosphate removal, the system allows for the simultaneous reduction of trace metals, such as cadmium, zinc or nickel, which are often found in UK municipal wastewater due to industrial effluent being discharged into the sewer.

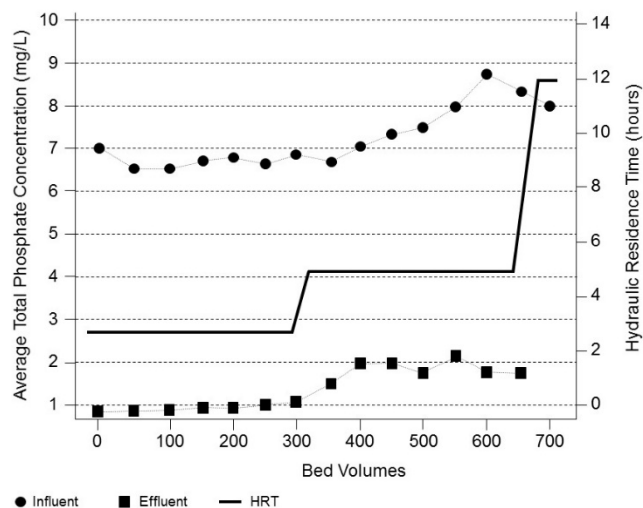
Where TP discharge consents of  $\leq 2.0\ \text{mg}/\text{L}$  are required, phosphate removal efficiency is approximately 80% per filter pass (i.e., per filter bed volume). Bed residence times are typically between four and 12 hours, depending on influent phosphate concentrations and discharge consents. Phosphate removal capacity is projected to be  $>14\ \text{g}$  of phosphate per kilogram of pellets. This capacity equates to bed (or filter) longevity of one to seven years, depending on influent TP levels, customer and site requirements, flow rates, site consent levels, and other system design criteria; “spent” or fully-charged pellets can be reused in agriculture, as they are “phosphate-enriched”.



**Fig 2:** Close-up image of the pellets used in this research study (left); SEM image of the interconnected pore space of pellets used in the technology (right).

Independent analyses of the technology have been conducted in Australia and the United Kingdom. For example, research conducted by the Water Research Council (WRC) in the UK investigated the kinetics, chemistry and outcomes of the technology for TP treatment (Water Research Council, 2005; Tillotson, 2006) [32, 30]. This research showed that while TP influent levels ranged from  $6.5\ \text{mg}/\text{L}$  to  $9.0\ \text{mg}/\text{L}$ , TP effluent levels were  $\leq 2.0\ \text{mg}/\text{L}$  when hydraulic residence time (HRT) was between four and 12 hours.

Figure 3 shows the relationship between influent TP levels versus effluent TP levels as a function of the number of bed volumes (i.e., the number of times effluent passed through the filter beds on a per bed volume basis) observed in the WRC study. This comparison is then matched to the time influent remained in contact with the pelletised media, represented as HRT. It was found that as HRT increased from three to five hours and from five to 12 hours due to elevating levels of influent TP, phosphate removal efficiencies remained the same, although effluent TP levels also predictably rose.



**Fig 3:** Influent and effluent TP concentrations and HRT for municipal wastewater.

These results suggest that up to 2,000 bed volumes of phosphate-rich effluent can be processed by one filter when used in an upward flow design. For such an application, HRT is approximately six hours (by way of comparison, reed bed applications may be as long as 24+ hours). In the WRc configuration, >80% TP was removed in the first filter pass, with a further 80% TP removed in each subsequent filter bed

pass. This study also indicated that phosphate breakthrough occurred. Figure 4 shows TP concentrations in both the influent and effluent as a function of time (bed volumes per hour), showing that effluent TP levels of  $\leq 2.0$  mg/L can be reached when influent TP levels average 8.0 mg/L. These influent TP levels are typical at the tertiary stage of treatment in most small- and medium-sized WTPs in the UK.

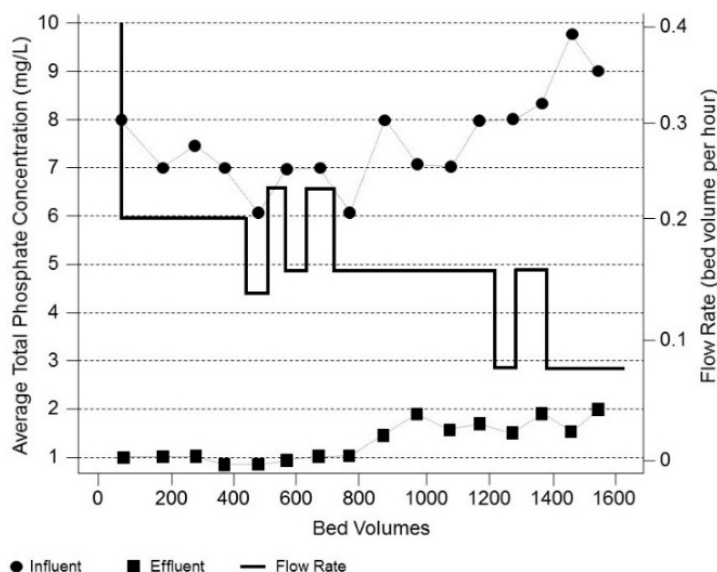


Fig 4: Influent and effluent levels before and after treatment when influent levels average 8.0 mg/L.

The WRc findings are largely consistent with those observed when a powdered form of the MARR reagent is applied directly to municipal wastewater after biological treatment (but before secondary clarification) based on data from research conducted in Australia (Fergusson, 2010) [21]. For example, Table 1 shows the results of applying the technology (dosing at a point between the bio-reactor and secondary clarifier) at Kilcoy Shire Council, a small municipal trickling-filter WTP

with an equivalent population (EP) of 2,000 and at Brisbane Water, a large AS WTP with an EP of 50,000+. Of note is the increased hydraulic throughput (created by improved settling times in secondary clarification), improvements in turbidity, colour and clarity, and the removal of objectionable odour at the Kilcoy WTP (although TN reductions are an anomaly not observed elsewhere).

Table 1: Influent and effluent levels before and after direct addition of the technology at Kilcoy Shire Council and Brisbane Water WTPs when compared to conventional treatment.

Parameter	Influent	After Conventional Treatment	After MARR Direct Addition	Percent Change
<i>Kilcoy Shire Council, Queensland, Australia</i>				
Hydraulic Throughput	400 kL/day	400 kL/day	600 kL/day	↑50%
BOD	500 mg/L	30 mg/L	9.0 mg/L	↓99%
TSS	200 mg/L	20 mg/L	1.0 mg/L	↓99%
TP	70 mg/L	14 mg/L	0.05 mg/L	↓99%
TN	70 mg/L	13 mg/L	3.9 mg/L	↓95%
E-Coli	26,000,000 cfu	10,000 cfu	7,000 cfu	↓99%
Total Metals	0.6 mg/L	0.6 mg/L	0.2 mg/L	↓60%
Colour	400 PCU	35 PCU	10 PCU	↓97%
Turbidity	240 NTU	16 NTU	1.0 NTU	↓99%
Clarity	20 mm	200 mm	2,200 mm	↑100%
pH	8.0-9.7	6.8-7.3	7.0-7.2	–
Objectionable Odour	Extremely High	High	Extremely Low	–
<i>Brisbane Water, Queensland, Australia</i>				
BOD	170 mg/L	3.1 mg/L	9.0 mg/L	↓95%
TSS	298 mg/L	9.9 mg/L	5.9 mg/L	↓98%
TP	7.2 mg/L	6.3 mg/L	1.9 mg/L	↓74%
TKN	47 mg/L	29 mg/L	29 mg/L	↓40%

However, given the proposed lower consent levels of TP in Europe, a thorough investigation of this technology for longer operational durations in the UK has not been carried out, and therefore the present research study asked the following

questions: 1) when configured as an upward vertical flow filtration system, does the technology reduce phosphate (measured as TP and total orthophosphate) to  $\leq 2.0$  mg/L when applied at a trickling filter WTP in the UK; 2) what HRTs are

required to answer research question 1); 3) does multi-stage filtration with a shorter HRT remove TP more effectively than single-stage filtration with a longer HRT; 4) what is the longevity of the filter media; 5) does the filtration system remove BOD, ammonia and heavy metals, and if so by what percentage; and 6) what are the estimated costs of the technology in relation to other forms of chemical treatment?

### Method

Yorkshire Water conducted a four-month independent assessment of the technology at its Kirk Smeaton trickling filter WTP, a small, rural, unmanned municipal waste treatment facility in West Riding, North Yorkshire. The design of the system comprised three separate filter configurations to compare different design scenarios, HRTs, pellet size fractions, removal efficiencies and projected filter life-spans.

**Filter Pairing A** consisted of two filters, each containing approximately 0.75 m<sup>3</sup> (equivalent to about 750 kg) of 10-25 mm pellet media. Each filter had an HRT of six hours for a total HRT of 12 hours. For four months, samples were taken from the influent point, an intermediate point between filters #1 and #2, and at discharge. This configuration, with labelled unit processes, is shown in Figure 5. Three composite samples were taken daily: one at the inlet of the filter system; one intermediate sample (at the point between each filter); and a treated effluent sample. The intermediate sample was taken from a sample point situated between the outlet of the first filter and the inlet of the second filter.

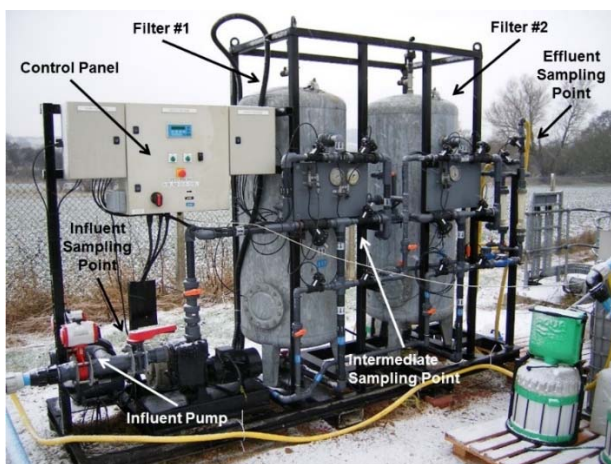


Fig 5: Unit processes of Filter Pairing A.

**Filter Pairing B** consisted of a two-stage filter running in series, with each filter holding approximately 20 kg of 5-10 mm pellets. The filters were initially operated with a one-hour residence time in each filter, giving a total HRT of two hours across the filter pairing. During the project, HRT was subsequently reduced to a total of one hour across the filter pairing (30 minutes in each filter) for a two-week period and then increased to a total of three hours across the filter pairing (one and half hours in each filter) for the remainder of the trial. Three composite samples were taken daily: one at the inlet of the filter system; one intermediate sample (at the point between each filter); and a treated effluent sample. The intermediate sample was taken from a sample point situated between the outlet of the first filter and the inlet of the second filter.

**Filter Pairing C** consisted of three filters, each filter had a residence time of four hours, giving a total HRT across the filters of 12 hours. The three filters, like Filter Pairing B, contained approximately 20 kg of 5-10 mm pellets. The filters were designed to replicate Filter Pairing A, except using three filters. Three composite samples were taken daily: one at the inlet of the filter system; two intermediate samples (intermediate points A and B between filters #1 and #2 and between filters #2 and #3); and a treated effluent sample. The first intermediate sample was taken from a sample point situated between the outlet of the first filter and the inlet of the second filter; the second intermediate sample was taken from a sample point situated between the outlet of the second filter and the inlet of the third filter.

### Results

**Filter Pairing A** had an average TP load of 10.3 mg/L in the influent, which resulted in a TP load of 5.2 mg/L at the intermediate point, and a final treated effluent TP of 2.6 mg/L, representing a 74% reduction in TP. On average, approximately 5.0 mg/L of phosphate was removed by the intermediate stage and 2.5 mg/L of phosphate was removed by the second (final effluent) stage. Data from Filter Pairing A can be seen in Figure 6. TP removal efficiencies mostly followed the same variations in influent TP loadings.

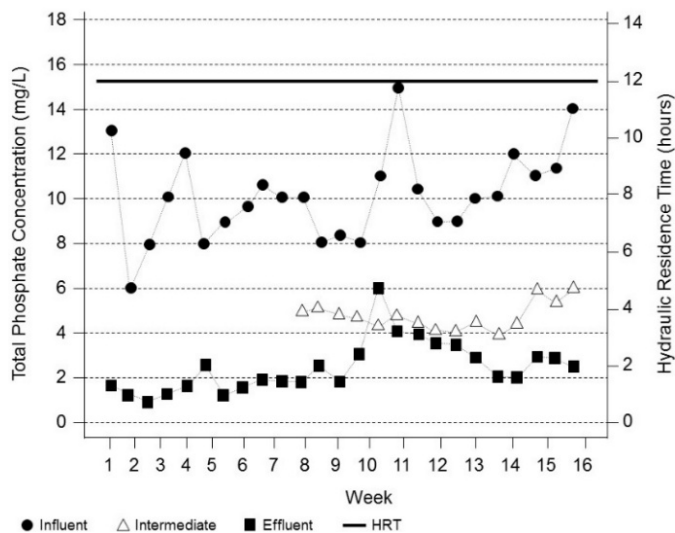


Fig 6: Influent and effluent TP levels and HRT times for Filter Pairing A.

Filter Pairing A also had a total orthophosphate (H<sub>3</sub>PO<sub>4</sub>-) load of 9.6 mg/L in the influent, which resulted in a total orthophosphate load of 4.6 mg/L at the intermediate point (between filters #1 and #2), and a total orthophosphate of 2.1 mg/L in the final treated effluent, representing a 77% reduction of total orthophosphate. Final effluent pH was on average 9.1. BOD was 6.8 mg/L in the influent, and 4.3 mg/L in the effluent, a 36% reduction; ammonia (NH<sub>3</sub>) concentrations were 1.1 mg/L in the influent, 1.0 mg/L at the intermediate point, and 0.86 mg/L in the effluent, a 22% reduction. Similarly, reductions in chromium (<0.01 mg/L reduced to 0.019 mg/L) and zinc (0.05 mg/L reduced to <0.02 mg/L) were observed.

**Filter Pairing B** had a TP load of 9.6 mg/L in the influent, which resulted in a TP of 3.6 mg/L at the intermediate point

(between filters #1 and #2), and a TP of 1.8 mg/L in the final treated effluent, representing an 81% reduction in TP. On average, approximately 6.0 mg/L of total phosphate was removed by the intermediate stage and about 2.0 mg/L of remaining phosphate was removed by the second filtration stage. Final effluent pH averaged 8.6, and data from Filter Pairing A can be seen in Figure 7.

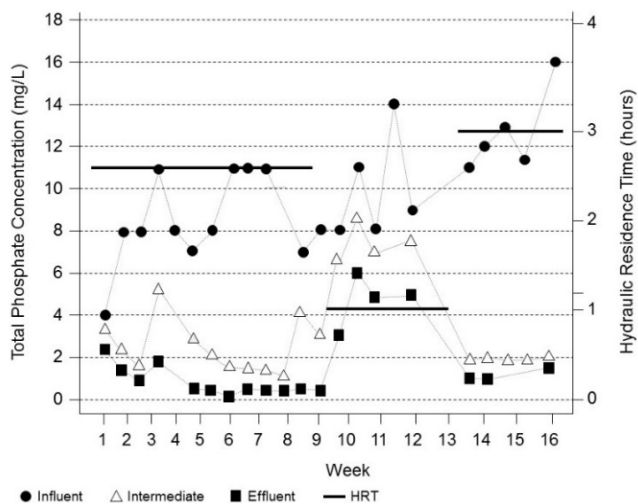


Fig 7: Influent and effluent TP levels and HRT times for Filter Pairing B.

**Filter Pairing C** had a TP load of 9.6 mg/L in the influent, which resulted in a TP load of 1.1 mg/L at the first intermediate point (between filters #1 and #2), a TP load of 0.5 mg/L at the second intermediate point (between filters #2 and #3), and a TP of 0.37 mg/L in the final treated effluent, representing a 96% reduction in TP. On average, approximately 8.5 mg/L of phosphate was removed by the first intermediate stage, 0.64 mg/L of phosphate was removed by the second intermediate stage, and 0.13 mg/L was removed by the third filtration stage; data from Filter Pairing C is shown in Figure 8, noting that TP only rose above 2.0 mg/L when influent TP rose above 13 mg/L to 16 mg/L in week 16.

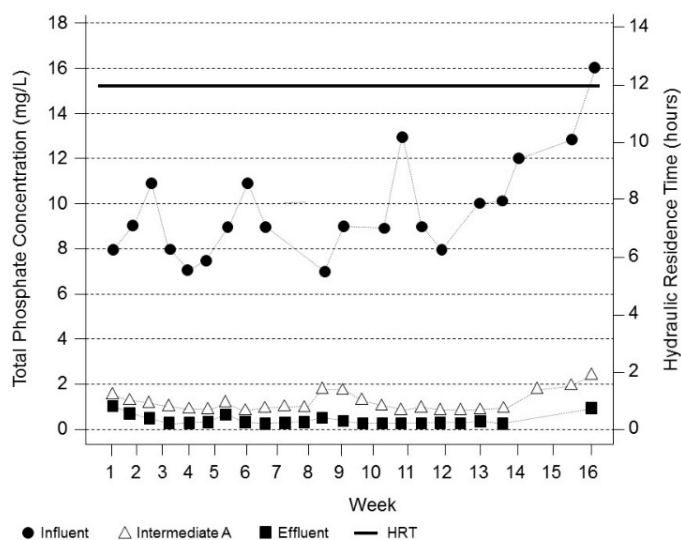


Fig 8: Influent and effluent TP levels and HRT times for Filter Pairing C.

The WRc research cited above demonstrated a loading capacity of approximately 6.0 g of phosphate per kg of pellets (or 6.0 kg/m<sup>3</sup>), although it calculated the filter columns used in its research would continue to remove TP to higher levels than those observed. However, earlier long-term laboratory trials indicated this may be as much as >14 g/kg (14 kg/m<sup>3</sup>) when pellets are exposed to phosphate for longer times. In both instances, loading rates were in excess of 90%. These levels of phosphate loading capacity are higher than those observed for alternate chemical dosing methods. For example, the Water Research Council (2005) [32] reported the following:

**Sand and Gravels:** According to the WRc, it has been estimated the phosphate adsorption capacity of a tertiary, horizontal-flow reed bed filled with the most efficient sand would be depleted in a period of approximately six months; for less efficient sands, TP removal would only continue for two months (Del Bubba *et al.*, 2003) [9]. For example, phosphate removal rates of 0.4 kg/m<sup>3</sup> for horizontal-flow and for vertical-flow reed beds are not uncommon. It has also been shown by Rustige *et al.* (2003) [29] that a study of the TP removal capacity of 62 sand-based, sub-surface horizontal-flow constructed wetlands in Germany, Austria and Switzerland indicated that while 50% of horizontal-flow units had an average TP output concentration of 2.1 mg/L, the vertical-flow units had an output concentration of 3.3 mg/L. These findings suggest that vertical-flow beds with an average operating time of five years were less effective than horizontal-flow units with an average operating time of 8.5 years; to achieve treatment levels of TP <1.0 mg/L, both reed bed designs proved inadequate. Heal *et al.* (2004) have shown that bottom ash has a phosphate removal capacity of 0.06 g/kg, steel slag 0.38 g/kg, blast furnace slag 0.40-0.45 g/kg, fly ash 0.62 g/kg, shale 0.75 g/kg, laterite 0.75 g/kg, and zeolite 1.0 g/kg, although different forms of ochre, while not readily available, can remove upwards of 26 g of phosphate per kilogram.

**Expanded Clay:** It has been shown that lightweight, expanded clay aggregates (known as Norwegian Leca/Filtralite) in reed beds have a TP removal capacity of about 0.5 kg/m<sup>3</sup> (Zhu *et al.*, 2003) [34]. Clay has the advantage over other mineral media that once phosphorus removal capacity has been reached and the media requires replacement, it can be used as an agricultural fertiliser if waste management licensing permits. This advantage also exists with the pelletised media used in this study, but the removal rates for clays are low and do not favourably compare with those observed herein.

**Calcite Media:** Tests indicate that calcite media (such as calcium carbonate, CaCO<sub>3</sub>) has a TP sorption capacity of approximately 2.0 kg/m<sup>3</sup> (Arias *et al.*, 2003) [1]. Arias *et al.* found the TP removal capacity of calcite filters when used operationally in a WTP was about tenfold lower than that found under laboratory conditions. They reported that the formation of bio-films on the calcite and short retention times in the filter may explain its reduced effectiveness for TP removal.

Using the results obtained from Filter Pairing A, the longevity of the system was estimated as follows: Assuming the media removes 14 g/kg, and Filter Pairing A holds a total of 1,500 kg, the filter had the potential to remove 21,000 g of phosphate. The total amount of phosphate removed over the duration of

the 16-week study was calculated to be 2,035 g, thus the filter system had the potential to remove a further 18,965 g. From this it was calculated that with a 12-hour HRT the average removal rate of TP over the duration of the study was 0.007 g/L. By calculating the amount of phosphate removed in a year, it was determined the system at Kirk Smeaton would remove phosphate at the recorded rate for 958 days, giving the filter a projected lifespan of a further 2.5 years. Utilising the same principle, with regard to applying the technology as polishing filter using an HRT of six hours, the estimated lifespan of the media was predicted to be approximately 9.5 years. As shown in Table 2, the cost per kilogram of TP removed from municipal wastewater using chemical dosing with ferric chloride is sensitive to the size of the WTP, and increases in cost are experienced when consent levels of  $\leq 2.0$  mg/L are

imposed (Environmental Data Services, 2006d) [16]. The operating cost of chemical dosing for small- (i.e., EP <2,000) to medium-sized (i.e., EP <10,000) WTPs is £15-£93 per kilogram of phosphate removed (at an average of £46 per kilogram of phosphate), depending on TP consent levels when heavy metal consents are not imposed, and £23-£146 per kilogram of phosphate removed (at an average cost of £72 per kilogram of phosphate), depending on TP consent levels when heavy metal consents are also imposed. From this data it can be seen that the cost increase to reduce TP from 2.0 mg/L to 1.0 mg/L is non-trivial, and even more significant increases in cost can occur when heavy metal consents are imposed. As such, standard forms of chemical dosing may not represent a cost-effective option for small- or medium-sized WTPs.

**Table 2:** A summary of costs associated with standard chemical dosing at WTPs in the UK.

Level of TP Consent and Size of WTP	Cost (£/kgPO <sub>4</sub> ) of Phosphate Removal Without Metals Consent	Cost (£/kgPO <sub>4</sub> ) of Phosphate Removal With Metals Consent
TP consent of <1.0 mg/L		
<i>Small WTP</i>	£93	£146
<i>Medium WTP</i>	£38	£60
TP consent of 1.0 mg/L		
<i>Small WTP</i>	£67	£105
<i>Medium WTP</i>	£27	£43
TP consent of 2.0 mg/L		
<i>Small WTP</i>	£35	£56
<i>Medium WTP</i>	£15	£23

Additionally, water companies are increasingly concerned that phosphate treatment chemicals (e.g., ferric salts) may become more expensive due to a limited supply, thereby driving up chemical dosing costs further as the demand for chemicals grows and traditional sources for such salts (from the steel industry, for example) become scarcer.

For biological treatment, a cost of £46 per kilogram of phosphate removed has been estimated for small WTPs, and a cost of £11 per kilogram of phosphate removed for WTPs with an EP 2,000-80,000+ has been predicted (Environmental Data Services, 2006d) [16]. Although this latter figure is generally less than chemical dosing, it still represents a significant cost for smaller WTPs, and typically does not result in the lower levels of TP removal required by the new standards (i.e.,  $\leq 2.0$  mg/L).

The technology researched in this study has a price bracket of £23-£27 per kilogram of phosphate removed at small- to medium-sized WTPs, and thus appears to be cost-competitive with chemical and biological dosing methods. Its waste-to-resource credentials and the potential downstream reuse of spent media may make it more attractive to municipal councils in the UK and elsewhere in Europe.

### Conclusion

The following conclusions can reasonably be drawn from this research findings:

**Finding a)** despite reducing TP by 74% and orthophosphate by 77%, Filter Pairing A did not reduce either to  $\leq 2.0$  mg/L, although both Filter Pairings B and C did, thereby answering research question 1) in the affirmative;

**Finding b)** in order to achieve the goal of  $\leq 2.0$  mg/L, the HRT for Filter Pairing B was two hours and for Filter Pairing C was 12 hours over three filters, thereby answering research question

2); **Finding c)** indicates that multi-stage filtration results in greater phosphate removal rates than single-stage filtration, thereby answering research question 3) in the affirmative; **Finding d)** suggests the estimated bed life of the filter media was between 2.5 and 9.5 years, depending on HRT and levels of phosphate to be treated, thereby answering research question 4); **Finding e)** the technology was effective in removing 35% of BOD, 25% ammonia and some heavy metals from municipal wastewater, thereby answering research question 5) in the affirmative; and

**Finding f)** the average operating cost to reduce TP to consent levels of  $\leq 2.0$  mg/L using ferric chloride is £65 per kilogram of phosphate for small-sized WTPs in the UK, compared to £25 per kilogram of phosphate using this technology, thereby answering research question 6).

The technology appears to be a sustainable wastewater treatment methodology. The “spent” media, once fully charged with phosphate, has the potential to be reused as a soil remediation treatment media, as a soil amendment agent, or as an additive to brick and concrete manufacture as a fluxing agent or colorant, among other potential reuse options (Fergusson 2015) [22]. Removal of the media from the filter or a retro-fitted lagoon or reed bed can be achieved by suction, and the system provides a robust, low-maintenance means of achieving phosphorus removal to low levels, with a competitive longevity of filter beds. The system was also easy to fit (or could be retro-fitted) to the existing treatment system. Moreover, the treatment system operated effectively without the need for further auxiliary chemical dosing or influent pH adjustment, thus removing the need for the handling and storage of hazardous chemicals such as FeCl<sub>3</sub>, and thereby reduced operator intervention requirements, cost and risk at the unmanned site. The average operating cost of phosphate

removed may make the technology cost-competitive for small- and medium-sized WTPs, with a cost differential (particularly over chemical dosing) for works where the population equivalent is <10,000.

However, several important research questions remain to be answered, including the potential impact of using 10-25mm pellets in Filter Pairing A versus 5-10mm pellets in Filter Pairings B and C. The significantly greater surface area of these smaller pellets may have contributed to the removal efficiencies observed here, and such side-by-side assessments of the role of size fractions must be investigated. Furthermore, the actual filter or bed life of such a system needs far more rigorous testing than the estimated lifespans advanced by this research. For the technology to be economically viable and therefore adopted as an alternative to ferric chloride dosing, a greater understanding of pellet longevity is required.

Finally, while this research highlights the waste-to-resource character of the pelletised media (i.e., the reuse of modified alumina refinery residue, an industrial waste as a source of pellet production and thus the treatment of TP in municipal wastewater), it does not go far enough in explaining its potential for further sustainable reuse. For example, once the pellets are spent or “wasted” as a result of this type of application, can they really be reapplied as a soil amendment additive, and if so what is their potential chemical, nutrient and/or commercial value? While research indicates that MARR itself may serve this purpose (e.g., Rai *et al.*, 2012) [28], spent pellets loaded with phosphate have yet to be so examined in the UK or Europe. The licensing requirements and limitations associated with applying industrial “waste” to soil must also be fully known and tested before such claims can be robustly defended.

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