1 New approaches to enhance pollutant removal in artificially

2 aerated wastewater treatment systems

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11 Abstract

13	Freshwater ecosystems sustain human society through the provision of a range of services.
14	However, the status of these ecosystems is threatened by a multitude of pressures, including point
15	sources of wastewater. Future treatment of wastewater will increasingly require new forms of
16	decentralised infrastructure. The research reported here sought to enhance pollutant removal
17	within a novel wastewater treatment technology, based on un-planted, artificially aerated,
18	horizontal subsurface flow constructed wetlands. The potential for these systems to treat de-icer
19	contaminated runoff from airports, a source of wastewater that is likely to grow in importance
20	alongside the expansion of air travel and under future climate scenarios, was evaluated. A new
21	configuration for the delivery of air to aerated treatment systems was developed and tested, based
22	on a phased-aeration approach. This new aeration approach significantly improved pollutant
23	removal efficiency compared to alternative aeration configurations, achieving > 90 % removal of
24	influent load for COD, BOD_5 and TOC. Optimised operating conditions under phased aeration were
25	also determined. Based on a hydraulic retention time of 1.5 d and a pollutant mass loading rate of

26 0.10 kg d⁻¹ m⁻² BOD₅, > 95 % BOD₅ removal, alongside final effluent BOD₅ concentrations < 21 mg L⁻¹, 27 could be achieved from an influent characterised by a BOD₅ concentration > 800 mg L⁻¹. Key controls on oxygen transfer efficiency within the aerated treatment system were also determined, revealing 28 29 that standard oxygen transfer efficiency was inversely related to aeration rate between 1 L and 3 L min⁻¹ and positively related to bed media depth between 1,500 mm and 3,000 mm. The research 30 31 reported here highlights the potential for optimisation and subsequent widespread application of 32 the aerated wetland technology, in order to protect and restore freshwater ecosystems and the 33 services that they provide to human society.

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35 Statement of Contributions: AF conceived the research, led the design and implementation of the experiments, analysed samples,

36 undertook statistical analysis and co-led the writing of the manuscript. BS contributed to the conception of the research and design of the

37 experiments and co-led the writing of the manuscript with AF. PH, MS and MM supported the design and implementation of the research

- 38 and edited earlier versions of the manuscript.
- 39

40 Key Words

- 41 Aerated constructed wetlands
- 42 De-icer contaminated runoff
- 43 Phased artificial aeration
- 44 Oxygen transfer efficiency
- 45 Organic pollutants
- 46 Freshwater ecosystems

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52 1. Introduction

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54 Freshwater ecosystems provide services that are critical for human society (Dodds et al., 2013, 55 UNESCO, 2015, Durance et al., 2016). However, these ecosystems also face diverse pressures 56 resulting from population growth, urbanisation, industrial development and a changing global 57 climate (Vorosmarty et al., 2000, Ormerod et al., 2010, Vorosmarty et al., 2010, Matthews, 2016). In 58 consequence, contemporary rates of degradation within freshwater ecosystems significantly exceed 59 historical rates, but also contemporary rates of degradation within other ecosystems (Barnosky et 60 al., 2011, Valiente-Banuet et al., 2015). The changes in ecosystem structure and function that are 61 associated with degradation threaten the integrity of freshwater ecosystems, but also constrain the 62 potential for human society to benefit from the services that could potentially be provided by 63 freshwaters (Gleick, 1998, World Health Organization, 2015). 64 Therefore, there is a growing imperative to protect and restore the status of freshwater

65 ecosystems globally. Chemical water quality is a fundmanetal control on ecosystem status and remains subject to significant anthropogenic pressure (Smith and Schindler, 2009, Schindler, 2012, 66 67 Malaj et al., 2014, Jekel et al., 2015, Van Meter et al., 2016). Point sources have been recognised as a 68 major contributor of pollutants to freshwaters for several decades in many countries (EEA, 2007, 69 DEFRA, 2012, EEA, 2015), with centralised or decentralised wastewater treatment systems being 70 widely used to improve the quality of wastewater entering freshwaters from point sources. 71 However, the energy demands, greenhouse gas emissions and whole life costs associated with many 72 traditional wastewater treatment technologies are subject to increasing scrutiny (Henriques and 73 Catarino, 2017, Rajasulochana and Preethy, 2016). Alternative treatment technologies, characterised 74 by relatively low energy consumption, by simple operating principles and by minimal whole life costs 75 are increasingly required. Such technologies provide a potentially more sustainable means of 76 protecting or enhancing ecosystem services compared to conventional wastewater treatment 77 technologies. Further, such technologies would support enhanced treatment of point sources in

78 countries where significant investment in centralised wastewater treatment infrastructure cannot be 79 made, alongside the treatment of smaller, micro-point sources of wastewater for which traditional 80 technologies may be inappropriate or disproportionately costly. In this context, the research 81 reported here developed a novel treatment technology for wastewater, based on un-planted, 82 artificially aerated, horizontal subsurface flow (HSSF) constructed wetlands. It is recognised that the 83 treatment systems considered in this research do not include the vegetation planting schemes that 84 are common in many constructed wetland designs. This reflects the specific application of the 85 technology to the treatment of wastewater at airports, for which planted systems are potentially 86 inappropriate (see below). However, the HSSF constructed wetland terminology is used within this 87 paper, reflecting the fact that in hydraulic, microbial and bed media geochemical terms, the 88 treatment systems described here share many features that are common with planted HSSF 89 constructed wetlands.

90 Treatment technologies that rely on natural, passive pollutant degradation processes, including 91 constructed wetlands, offer environmental and economic advantages over many traditional 92 wastewater treatment systems (Castro et al., 2005, Vymazal et al., 2006, Kadlec and Wallace, 2009, 93 Vymazal and Kröpfelová, 2009, Freeman et al., 2015, Wu et al., 2015). However, the availability of 94 dissolved oxygen (DO) is frequently a fundmanetal limit on pollutant removal within traditional 95 constructed wetland designs (Wallace et al., 2007, Kadlec and Wallace, 2009, Nivala et al., 2013). In saturated HSSF wetlands, 0.12 g m⁻² d⁻¹ – 12.11 g m⁻² d⁻¹ O₂ may be transported into a system 96 97 through the combination of direct diffusion from the atmosphere and diffusion from the sub-surface 98 root network of wetland vegetation (Armstrong et al., 1990, Brix and Schierup, 1990, Brix, 1997, 99 Bezbaruah and Zhang, 2005, Nivala et al., 2013). Vertical flow constructed wetlands achive higher 100 diffusion rates of 28.4 g m⁻² d⁻¹ – 156 g m⁻² d⁻¹ O₂ for saturated systems and up to 482 g m⁻² d⁻¹ O₂ in 101 fill and drain systems, primarily due to the draw down of air into the bed during sequential filling and 102 draining of wastewater through the wetland substrate (Cooper, 2005, Fan et al., 2013a, Nivala et al., 103 2013). However, the rate of DO supply via these mechanisms within traditional constructed wetland

104 designs is often negligible when compared to the rate of DO consumption associated with many raw 105 wastewaters (Nivala et al., 2013). Whilst anaerobic respiration of some pollutants occurs, the 106 resulting pollutant removal rates are often lower than under aerobic conditions, meaning that 107 treatment efficiency is significantly reduced (Huang et al., 2005, Ouellet-Plamondon et al., 2006, Fan 108 et al., 2013b, Nivala et al., 2013, Murphy et al., 2015, Uggetti et al., 2016). The need to improve rates 109 of DO supply in order to enhance pollutant removal in traditional constructed wetlands has led to 110 the development and commercialisation of aerated wetlands for a range of applications across the 111 globe (Wallace, 2001). Aerated wetlands involve the active supply of DO into a self-contained, 112 media-filled treatment bed, to maintain aerobic conditions within the wetland by meeting the DO 113 demand exerted by wastewater. With sufficient DO supplied to the system through aeration, the 114 role of wetland vegetation root transfer for this purpose is significantly reduced and systems can 115 remain un-planted to serve applications in which attacting wildlife is undesirable. However, there is 116 currently no recognised design standard for aerated wetlands (Nivala et al., 2013), alongside limited 117 empirical data to support understanding of how factors such as the configuration of aeration 118 devices, wetland bed depth or aeration rate impact the availability of DO and, ultimately, pollutant 119 removal. In alternative aerobic wastewater treatment systems, such as activated sludge plants, the 120 energy consumed by aeration devices typically accounts for 45 % to 80 % of the total operating cost 121 (Stenstrom and Rosso, 2006, Zhou et al., 2013). Optimisation of aeration systems within wastewater 122 treatment plants can typically achieve energy efficiency improvements of 20 % to 40 % (Henriques and Catarino, 2017). Therefore, the design and optimisation of aeration systems to ensure maximum 123 124 O₂ transfer from the gaseous to the liquid phase is central to achieving low cost, sustainable 125 treatment solutions through the use of aerated wetlands.

The research reported here developed and tested novel, artificially aerated HSSF constructed wetland designs to enhance pollutant removal efficiency from wastewater. The specific context for the research was the need for new technological approaches to treat surface water runoff from airports following contamination with chemical de-icers. At international hub airports, > 1 M L of

130 chemical de-icers are applied annually to aircraft, aprons, taxiways and runways to facilitate safe 131 winter operations, potentially contaminating large volumes of storm water runoff (CAA, 2000, ACRP, 132 2008, Erdogan, 2008, Association of European Airlines, 2012, ISO, 2012, Freeman et al., 2015). De-133 icer application volumes at UK airports are forecast to increase in-line with an increase in aircraft 134 movements of 1 % to 3 % annually up to 2050 (DFT, 2003, DFT, 2013). Simultaneously, expansion of 135 airports to meet this demand alongside a changing global climate will likely generate increasing 136 volumes of surface water runoff, placing unprecedented pressure on existing wastewater 137 infrastructure and pollution prevention strategies, potentially threatening the status of freshwater 138 ecosystems.

139 The primary environmental concern associated with airport de-icing activities is DO depletion in 140 surface waters that receive storm water runoff, due to the high DO demand exerted by propylene 141 glycol (C₃H₈O₂) or potassium acetate (C₂H₃KO₂) contained within chemical de-icers (ACRP, 2008, 142 ACRP, 2009). Both $C_3H_8O_2$ and $C_2H_3KO_2$ are associated with extremely high five-day biochemical oxygen demand (BOD₅) and, when diluted within snow melt or rainfall, can potentially achieve BOD₅ 143 144 concentrations > 20,000 mg L^{-1} within storm water runoff (US EPA, 2000, Corsi et al., 2012). Coupled with high temporal variability in pollutant and hydraulic loads, de-icer contaminated storm water 145 146 runoff from airports represents a significant threat to freshwater ecosystems if discharged without 147 appropriate treatment (Corsi et al., 2001, ACRP, 2008, ACRP, 2012).

148 De-centralised treatment of de-icer contaminated storm water runoff at airports using aerated 149 constructed wetlands represents a novel and potentially sustainable approach to the treatment of 150 this source of wastewater. Artificially aerated constructed wetlands appear to offer several 151 advantages in the context of treating de-icer contaminated runoff compared to conventional 152 wastewater treatment technologies, including: simple design; low maintenance; low operational and whole life costs; no regular sludge disposal requirements and resilience to fluctuating hydraulic and 153 154 pollutant loads (Kadlec and Wallace, 2009, Freeman et al., 2015). The limitations of artificially 155 aerated wetlands for airport applications include the large footprint required to manage large storm

156	water runoff volumes and the attraction of wildlife to wetland habitats which can pose a possible
157	bird strike hazard for aircraft (Blackwell et al., 2008). Despite such limitations, many airports have
158	large areas of land in close proximity to runways that could represent suitable sites for un-planted
159	HSSF aerated wetlands (Wallace and Liner, 2011a, Wallace and Liner, 2011b). In order to inform the
160	application of artificially aerated wetlands at airports, the objectives of the research reported here
161	were: (i) to evaluate the impact of media depth and aeration rates on standard oxygen transfer
162	efficiency (SOTE); (ii) to determine the impact of different aeration diffuser configurations on
163	pollutant removal efficiency; and (iii) to determine optimal pollutant loading rates for effective
164	treatment of de-icer contaminated runoff using aerated wetlands.
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166	2. Methodology and Materials
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168	2.1. Standard Oxygen Transfer Efficiency Tests
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170	Tests were conducted to determine standard oxygen transfer efficiency within four experimental
171	columns filled with 10 mm to 20 mm washed angular limestone gravel, to depths of 1,500 mm, 2,000
172	mm, 2,500 mm and 3,000 mm (Figure 1). Each individual column was constructed from 220 mm
173	internal diameter medium density polyethylene gas pipe and included sample ports, sample valves,
174	ceramic disc diffusers and airlines. The columns were sealed at the base with electrofusion couplings
175	and bolted stainless steel end caps, with drain valves and 200 mm fine bubble ceramic disc diffusers
176	positioned near to the base. Air was delivered into the columns using an Airmaster model 8/36, 1.5
177	hp, 24 L oil-free compressor. A 0 – 15 L min ⁻¹ flow meter was positioned on the compressor outlet
178	and used to regulate the aeration rate delivered into each column. Paired 10 mm internal diameter
179	sample ports were installed on opposite sides of the columns at elevations from the column base









Standard oxygen transfer efficiency tests were conducted following procedures described within the ASCE standard (ASCE, 2007). Briefly, DO concentration profiles were generated for each test, which first involved purging nitrogen gas through the column to deoxygenate the potable water within the media pore spaces in the column to < 0.5 mg L⁻¹ DO (Ghaly and Kok, 1988). The time taken for DO concentrations to reach the steady-state saturation point during re-aeration via an aeration device at a pre-calibrated flow rate was subsequently measured (Figure 2). The DO concentration data from the point of re-aeration to steady state saturation were analysed using the
ASCE-approved DOPar3-0-3 programme non-linear regression model and standardised to conditions
of 20 °C water temperature and 1,000 mbar barometric pressure (Stenstrom et al., 2006, ASCE,
2007).

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Figure 2. Example dissolved oxygen (DO) profile demonstrating the changing DO concentration
during standard oxygen transfer efficiency tests. Example shows the DO profile at sample location S1
(see Figure 1) within the 3,000 mm deep column.

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218 Dissolved oxygen concentrations were measured at ten-second intervals simultaneously at the 219 four sample locations (S1 - S4) using optical multi-parameter probes installed within flow cells 220 (Figure 3). Water was pumped through each flow cell at a rate of approximately 16 ml min⁻¹ using a 221 peristaltic pump to create a sealed, self-contained sample loop through which the test water was 222 continuously circulated. Pump tubing was purged prior to each test to remove any trapped air 223 resulting from filling of the column or calibration of the probes. Each probe was calibrated prior to each test following a two-stage calibration procedure for DO, involving the atmospheric saturation 224 225 point followed by a zero-point calibration within a 1000 mg L^{-1} Na₂SO₃ and 1 mg L^{-1} CoSO₄ solution. Three aeration rates (1 L min⁻¹, 2 L min⁻¹ and 3 L min⁻¹) were tested with each of the four media 226

227 depths, to assess the impact of aeration rate and total media depth on SOTE (Table 1). Tests were 228 conducted between 15/11/2014 – 05/01/2015, with each combination of media depth and aeration 229 rate repeated in triplicate. Temperature, DO, redox potential and total dissolved solids 230 concentration were recorded within the potable water prior to the start of each test, confirming that no significant changes in the quality of the potable water occurred between individual tests. 231 232 233 234 Media-filled column Bluetooth data logger 235 Low flow peristaltic pump 236 Multi-parameter probe installed Sample port 237 within flow cell 238 Flow direction (closed flow loop) 239 240 Figure 3. Photograph of an experimental column and the low flow recirculation pump, Smartroll ™ 241 242 RDO [®] multi-parameter probe and flow cell setup to ensure a closed, self-contained flow loop. This setup was replicated at each of the four sample locations during testing to obtain representative DO 243 244 concentrations at different depths within the column. 245 246 247 248 249 250

Test No.	Total media depth (mm)	Aeration rate (L min⁻¹)
1	1,500	1
2	1,500	2
3	1,500	3
4	2,000	1
5	2,000	2
6	2,000	3
7	2,500	1
8	2,500	2
9	2,500	3
10	3,000	1
11	3,000	2
12	3,000	3

Table 1. Experimental design for standard oxygen transfer efficiency tests in media-filled columns with total depths of 1,500 mm to 3,000 mm, operating at aeration rates of $1 L \text{ min}^{-1}$ to $3 L \text{ min}^{-1}$

252 2.2. Pilot-Scale Aerated Wetland Tests

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Aeration configuration and optimisation tests were conducted within an un-planted, pilot-scale 254 255 system that closely replicated an artificially aerated HSSF constructed wetland (Figure 4), located at 256 Manchester Airport, UK (53.356235 °N -2.282445 °W). The pilot-scale system comprised a 1,000 L 257 mixing tank and three cylindrical tanks (1,600 mm deep x 1,400 mm diameter) each of 2,500 L 258 capacity, replicating three aerated wetland treatment cells. A Marlow Watson 520R peristaltic pump 259 was used to dose wastewater from the mixing tank into the first treatment cell. The three treatment 260 cells were positioned in series and connected with 50 mm internal diameter flexi-hose. The elevation 261 of each successive cell decreased by 250 mm, allowing gravity to drive water flow through the 262 treatment system. Each cell comprised a non-insulated narrow inlet distribution zone containing 40 263 mm to 100 mm diameter crushed brick and a main treatment zone containing the same 10 mm to 20 264 mm diameter angular limestone gravel media used within the SOTE tests described in Section 2.1. Media within the main treatment zone of each cell was separated from the inlet zone by a slotted 265 266 mesh screen. As an alternative to wetland vegetation, the media was capped with a porous membrane and a 200 mm deep layer of bark chippings to provide insulation. The total media depth 267

268 within each cell was 1,400 mm, corresponding to a total media volume of 6.45 m³ across the three 269 treatment cells. Three 30 mm internal diameter piezometers with 50 mm long screens at the base were installed within each treatment cell to depths of -250mm, -750mm and -1,250mm below the 270 gravel media surface, enabling measurement of physicochemical conditions within each cell. A 210 w 271 272 Charles Austen ET200 linear diaphragm blower was used to deliver up to 200 L min⁻¹ (45 L min⁻¹ m⁻³ 273 of media) of air into the system at 0.15 bar of pressure. Braided PVC airlines of 10 mm internal 274 diameter connected the blower to uniformly distributed tubular fine bubble membrane diffusers, 275 which were positioned at the base of each cell. A manifold system was fitted to the aeration line to 276 control the delivery and spatial distribution of aeration volumes into each treatment cell. Prior to 277 undertaking each individual aeration configuration and optimisation test, the system was 278 conditioned for twice the hydraulic retention time (HRT) using the test mass loading rate (MLR) to promote steady-state conditions and microbial acclimatisation. Each aeration configuration and 279 280 optimisation test was repeated in triplicate.

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Figure 4. Cross-section of the un-planted pilot-scale system, replicating an artificially aerated
horizontal subsurface flow constructed wetland, used for aeration configuration and optimisation
tests.
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- 290 Synthetic influent was created to replicate typical chemical oxygen demand (COD), BOD₅ and
- 291 total organic carbon (TOC) concentrations within winter storm water runoff from airports. The
- 292 synthetic influent comprised base-flow runoff from the airfield catchment at Manchester Airport
- 293 containing minimal masses of de-icers and consequently relatively low mean background pollutant
- 294 concentrations (Table 2).
- 295

Table 2. Characteristics and composition of baseflow runoff for Manchester Airport's airfieldcatchment and aircraft (ADF) and pavement (PDF) de-icers which were combined to create thesynthetic influent used within the aerated wetland tests

Parameter	Unit	Base-flow runoff (a)	ADF ^(b)	PDF ^(c)	Experiment synthetic influent		
Farameter					L	м	н
BOD	mg/l	52.4	354,000	270,000	831	1,355	1,853
COD	mg/l	109	834,000	330,000	1,206	2,405	3,404
тос	mg/l	40.6	-	-	424	1,104	1,534
TSS	mg/l	21.9	-	-	61	123	162
рН		7.6	7	10.6	7.1	6.8	7

^(a) Data recorded from samples collected from Manchester Airport's airfield catchment 12/11/2014 - 27/03/15 during baseflow conditions, defined as catchment discharge volume < 1.00 L sec⁻¹ (n = 123) ^(b) Values for neat Kilfrost ABC-S Plus aircraft de-icing fluid recorded from material data safety sheet. Active ingredient is propylene glycol ^(c) Values for neat Safegrip pavement de-icing fluid recorded from material data safety sheet. Active ingredient is potassium acetate plus corrosion inhibitors. L, M, H = low, medium and high respectively, defined as 0.2%, 0.3% and 0.4% volume of de-icer: volume of baseflow runoff water respectively (-) Data unknown or not applicable.

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297 Other contaminants potentially present within the baseflow runoff include surfactants, solvents, 298 triazoles, polycitric aromatic hydrocarbons (PAHs), aldehyde, benzine, volatile organic compounds 299 (VOCs) and sulphates, which are deposited within airport catchments during standard airport 300 operations such as aircraft and ground vehicle washing and maintenance, refuling and combustion 301 of aviation fuels (Sulej et al., 2011, Sulej et al., 2012). Whilst these contaminants were not directly 302 measured in the research reported here, it is assumed that only low concentrations would have 303 been present within the baseflow runoff because mobilisation and transport from within airport 304 catchments mainly occurs during storm water runoff events (Sulej et al., 2012). Baseflow runoff was

305	spiked with aircraft and pavement de-icing fluids sourced from Manchester Airport and used widely
306	within the aviation industry in order to create the synthetic influent used within the aerated wetland
307	tests. Three individual influent concentrations were created, defined as low (L), medium (M) and
308	high (H) strength, containing 0.2 %, 0.3 % and 0.4 % volume of de-icer:volume of runoff respectively
309	(Table 2). A nutrient solution of urea and ammonium phosphate was added to the synthetic influent
310	at concentrations consistent with nutrient requirements for optimal microbial growth (Grady et al.,
311	1999, Wallace and Liner, 2010, Wallace and Liner, 2011a). The approximate ratio of BOD $_5$:N:P was
312	kept constant by adjusting the volume of the supplementary nutrient solution in relation to the
313	BOD₅ MLR for a test, ensuring that microbial processes were not constrained by N or P availability.
314	The volume of influent dosed into the system for each individual test was adjusted in order to
315	maintain the desired HRT within the three treatment cells.
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317	2.2.2. Aeration Configuration Tests within Pilot-Scale Aerated Wetland
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319	Four aeration configurations were tested to establish their impact on pollutant removal
320	efficiency: phased aeration (PA), uniform aeration (UA), inlet-only aeration (IA) and no aeration (NA).
321	The individual configurations were achieved by adjusting the manifold system to alter the spatial
322	distribution and volume of air delivered into each of the three cells (Table 3). Each of these tests was
323	dosed with the L strength influent with a mean BOD ₅ concentration of 810 \pm 60 mg L ⁻¹ and an areal
324	MLR of 0.09 \pm 0.01 kg d ⁻¹ m ⁻² BOD ₅ , alongside a HRT of 1.5 d across all three cells taken together.
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Aeration HRT ^(a)		BOD ₅	BOD₅ mass	Air vol. (L min⁻¹)	min ⁻¹) distribution across the system		
Configuration	(days)	(mg L ^{−1})	(kg d ⁻¹ m ⁻²)	Cell 1	Cell 2	Cell 3	
Phased (PA)	1.5	834	0.1	100	66.6	33.3	
Uniform (UA)	1.5	727	0.08	66.6	66.6	66.6	
Inlet-only (IA)	1.5	812	0.09	200	0	0	
None (NA)	1.5	868	0.1	0	0	0	
Mean		810 ± 60	0.09 ± 0.01				

Table 3. Summary of hydraulic retention time (HRT), five-day biochemical oxygen demand (BOD₅) influent concentration, areal mass loading rates and air volume distribution throughout the pilot system during aeration configuration tests

^(a) Hydraulic retention time across all three cells taken together calculated following Equation 3. ± 1 standard deviation of the mean.

330 2.2.3. Optimisation Tests within Pilot-Scale Aerated Wetland

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332	Further to the a	eration configuration	tests nine se	narate tests were	conducted between
332	i ultilei to tile a	eration configuration	i tests, iiiie se	parale lesis were	conducted between

333 17/02/2015 – 14/06/2015 to determine the effect of wetland operating conditions on pollutant

removal (Table 4). The novel PA configuration, as opposed to UA, IA or NA described in Table 3, was

maintained throughout these optimisation tests. Three different HRTs (2.2 d, 1.5 d and 1.1 d) across

all three cells taken together were tested to assess the impact of HRT on pollutant removal

337 efficiency. Each of the three HRTs was tested with L, M and H strength influent as described within

338 Section 2.2.1., with respective BOD₅ concentrations of 831 \pm 35 mg L⁻¹, 1,355 \pm 81 mg L⁻¹ and 1,853 \pm

339 99 mg L⁻¹. During optimisation tests, operating conditions were equivalent to mean areal MLRs of

340 0.07 to 0.28 kg d^{-1} m⁻² BOD₅, within the typical range of areal MLRs (0.05 to 0.28 kg d^{-1} m⁻² BOD₅)

identified from the literature for uniformly aerated wetlands (Envirodynamics Consulting, 2012,

342 Moshiri, 1993).

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Test No.	HLR (m³ d⁻¹)	HRT ^(b) (days)	BOD₅ ^(c) (mg L ⁻¹)	BOD₅ (kg d ⁻¹ m ⁻²)
1	1	2.24	864 (L)	0.07
2	1.5	1.49	834 (L)	0.10
3	2	1.12	795 (L)	0.12
Mean (tests 1 - 3)			831 ± 35	0.10 ± 0.03
4	1	2.24	1,286 (M)	0.10
5	1.5	1.49	1,444 (M)	0.17
6	2	1.12	1,335 (M)	0.21
Mean (tests 4 - 6)			1,355 ± 81	0.16 ± 0.05
7	1	2.24	1,812 (H)	0.14
8	1.5	1.49	1,967 (H)	0.23
9	2	1.12	1,782 (H)	0.28
Mean (tests 7 - 9)			1,853 ± 99	0.22 0.69

Table 4. Summary of operating conditions including hydraulic loading rate (HLR), hydraulic retention time (HRT), five-day biochemical oxygen demand (BOD₅) influent concentration and influent areal mass loading rates used during aerated wetland optimisation tests one to nine ^(a)

^(a) Tests conducted with a phased aeration (PA) configuration and aeration rate of 44.64 m³ d⁻¹ m⁻³ of media across all three cells taken togeather,

^(b) hydraulic retention time across all three cells taken together calculated following Equation 3,

^(c) five-day biochemical oxygen demand (BOD₅) influent concentrations interpreted as L = low, M = medium and H = high strength. ± 1 standard deviation of the mean.

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350 2.2.4. Data and Sample Collection for Pilot-Scale Aerated Wetland Tests

352	Mean DO concentrations were determined by measuring DO in samples collected from the
353	piezometers installed within each cell on three occassions: at the start; mid-point; and the end of
354	each individual test. Dissolved oxygen was measured using a Hannah 9828 multi-parameter probe
355	within a sealed flow cell, through which approximately 16 ml min ⁻¹ of sample was pumped from the
356	piezometers using a peristaltic pump. Results were recorded when the probe readings had
357	stabilised, following purging of stagnant water from each piezometer. Further, a total of four spot
358	samples were collected for each individual aeration configuration and optimisation test, involving
359	one sample of the influent and one sample from each of the three treatment cell outlets. Samples
360	from the cell outlets were taken based on calculations of the HRT within each cell and assuming
361	steady state conditions and laminar throughflow within each treatment cell. Samples were collected

either manually into new, one litre plastic bottles, or via Aquacell P2 portable water samplers whichwere connected to the treatment cell outlets.

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365 2.2.5. Chemical Analysis of Water Samples from Pilot-Scale Aerated Wetland Tests366

367 Water samples from the aeration configuration and the optimisation tests were transported to 368 an on-site laboratory and analysed within 48 hours of collection for BOD₅, COD and TOC using 369 standard laboratory procedures. Several duplicate samples were also analysed by an independent 370 UKAS-accredited laboratory for BOD₅ following the ISO/IEC 17025 standard to verify the BOD₅ data 371 that were determined on-site. Digestion of samples on-site for COD and TOC was performed using a 372 LT200 instrument followed by colorimetric determination using a DR2800 photospectrometer. Hach 373 methods were used to standards ISO 6060-1989 for COD and EN 1484 purging method for TOC. Hach Addista LCA standards of 50 \pm 4 mg L⁻¹ COD and 16.5 \pm 3 mg L⁻¹ TOC were used to verify COD and 374 TOC results for each batch of samples processed. The analytical limit of detection was 15 mg L⁻¹ and 375 376 3 mg L⁻¹ for COD and TOC respectively. All samples outside of the method range of 3 mg L⁻¹ – 150 mg 377 L⁻¹ COD and 1.5 mg L⁻¹ – 30 mg L⁻¹ TOC were discarded and repeated following dilution with deionised water. Analysis for BOD₅ was performed at 20°C using a BODTrak ™ II instrument. Samples 378 379 were inoculated with a seed solution (PolySeed®) prior to incubation and analysed in accordance to 380 the Hach standard manometric sample dilution, five day test procedure method 8043 (Hach, 2013). 381 Blanks comprising de-ionised water, one nutrient buffer pillow and 35 ml of seed solution were frequently tested and discarded if the BOD₅ concentration was > 0.2 mg L⁻¹. In addition to external 382 383 laboratory verification, results for BOD₅ were verified on-site using glucose and glutamic acid (GGA) 384 standards of 300 mg L⁻¹, inoculated with 35 ml of PolySeed[®] solution and incubated at 20 °C for five 385 days following appropriate dilution. All GGA standard results were within the maximum standard 386 deviation of the method (\pm 30.5 mg L⁻¹).

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390	Pollutant removal efficiency for the tests described in Sections 2.2.2 and 2.2.3 was calculated as	s a
391	cumulative percent removal, R, between the influent and the final effluent leaving the third	
392	treatment cell following Equation 1, assuming that the system was in equilibrium at the time of	
393	sample collection:	
394		
395	$R = \frac{C_{\rm i} - C_{\rm o}}{C_{\rm i}} * 100$	(1)
396	where:	
397	R = pollutant removal efficiency (%)	
398	C_i = mean influent concentration across triplicate tests (mg L ⁻¹)	
399	$C_{\rm o}$ = mean final effluent concentration across triplicate tests (mg L ⁻¹)	
400		
401	Areal mass pollutant loading rates (kg d^{-1} m ⁻²) were calculated in accordance with Equation 2	
402	(Kadlec and Wallace, 2009):	
403		
404	$MLR = \frac{Q * C_i}{A}$	(2)
405	where:	
406	MLR = mass pollutant loading rate (kg d ⁻¹ m ⁻²)	
407	Q = volumetric flow rate (m3 d-1)	
408	C_i = influent pollutant concentration, i.e. BOD ₅ (mg L ⁻¹)	
409	A = wetland area (m^2)	
410		
411	Hydraulic retention time (HRT) was calculated in accordance with Equation 3 (Çakir et al., 2015,	
412	Metcalf and Eddy Inc, 1991):	
413		

414	$HRT = \frac{\pi r^{2} \ast \Phi \ast d}{Q}$	(3)
415	where:	
416	<i>HRT</i> = hydraulic retention time (days)	
417	π = pi	
418	r = cell radius (m)	
419	Φ = media porosity (40 %)	
420	d = media depth (m)	
421	Q = influent flow rate (m3 d-1)	
422		
423	2.3. Statistical Analysis	
424		
425	Two-way analysis of variance (ANOVA) and Tukey's-b tests were used to test the effects of me	edia
426	depth and aeration rate on SOTE within the experimental column tests. One-way ANOVA and	
427	Tukey's-b tests were performed on sample data from the aeration configuration tests to assess the	ıe
428	effects of aeration configuration within the system on COD, BOD_5 and TOC removal efficiency.	
429	Separate two-way ANOVA and Tukey's-b tests were performed on data from the aerated wetland	ł
430	optimisation tests to assess the effects of HRT and influent strength on COD, BOD₅ and TOC remo	val
431	efficiency. Data normality and homogeneity of variances were determined by Shapiro-Wilk and	
432	Levenne tests respectively, revealing normal distributions and variances to be homogeneous with	ווח
433	all datasets. All statistical analyses were conducted using IBM SPSS 20 and significant effects were	5
434	accepted at $p < 0.05$.	
435		
436		
437		
438		

439 3. Results

440

441 3.1. Effects of Aeration Rate and Media Depth on Standard Oxygen Transfer

442 Efficiency in Media-filled Columns

443

444	Aeration rate was inversely related to SOTE ($F(2,24) = 28.13$, MSE = 14.10, $p \le 0.0001$). Post-hoc
445	Tukey's-b tests revealed that aeration rates of 1 L min ⁻¹ resulted in significantly higher SOTEs
446	compared to aeration rates of 3 L min ⁻¹ . No significant difference was found between SOTEs under
447	aeration rates of 1 L min ⁻¹ compared to 2 L min ⁻¹ or aeration rates of 2 L min ⁻¹ compared to 3 L
448	min ⁻¹ . The effect of media depth on SOTE was not significant, nor was there a significant interaction
449	effect between aeration rate and media depth on SOTE. Whilst there was no significant effect of
450	media depth on SOTE, a consistent trend was observed with increasing media depth resulting in an
451	increase in mean SOTE under each of the three aeration rates (Figure 5).

452



Figure 5. Standard oxygen transfer efficiency (SOTE) in media-filled columns of 1,500 mm to 3,000
mm depth at aeration rates of 1 L min⁻¹ to 3 L min⁻¹. Columns represent the mean SOTE for each
depth and each aeration rate tested (n = 3 for each combination of aeration rate and media depth).



458 3.2. Impact of Aeration Configuration and Sample Position on Dissolved Oxygen

459 Concentration within the Pilot-Scale Aerated Wetland

460

461	The effect of aeration configuration on mean DO concentrations within pore water in the pilot-
462	scale aerated wetland cells was significant ($F(3,24) = 84.19$, $MSE = 2,158$, $p \le 0.0001$). Post-hoc
463	Tukey's-b tests indicated that the PA configuration resulted in significantly higher mean DO
464	concentrations within the treatment cells in comparison to UA, IA or NA configurations, whilst there
465	was no significant difference in mean DO concentrations between UA, IA and NA configurations. The
466	effect of sample position (cell number) on DO concentration was also significant ($F(2,24) = 57.19$,
467	<i>MSE</i> = 1,466, $p \le 0.0001$). Post-hoc Tukey's-b tests indicated that no significant difference in DO
468	concentrations was observed between cells one and two, however DO concentrations were
469	significantly higher in cell three compared with cells one and two. The interaction between aeration
470	configuration and sample position also had a significant effect on DO concentration ($F(6,24) = 45.61$,
471	<i>MSE</i> = 1,169, $p \le 0.0001$). Post-hoc Tukey's-b tests revealed that significant differences in DO
472	concentrations across the three cells were observed in PA and UA tests, but not within either IA and
473	NA tests in which DO concentrations remained at 0 mg L^{-1} across all three cells (Table 5).
474	

Table 5. Summary of mean dissolved oxygen (DO) concentrations (mg L^{-1}) determined from piezometer samples within treatment cells one to three during aeration configuration tests. n = 9 for each aeration configuration and cell position combination

Aeration	Po	osition within sy	stem
configuration	Cell 1	Cell 2	Cell 3
Phased (PA)	0	2.1 ± 1.2	8.3 ± 0.2
Uniform (UA)	0	0.7 ± 0.4	0.8 ± 0.4
Inlet-only (IA)	0	0	0
None (NA)	0	0	0

475

± 1 standard deviation of the mean

476 3.3. Impact of Aeration Configuration and Sample Position on Pollutant Removal

477 within the Pilot-Scale Aerated Wetland

478

479 Aeration configuration had a significant effect on COD (F(3,24) = 327.57, MSE = 3,403, $p \le$ 480 0.0001), BOD₅ (F(3,24) = 361.21, MSE = 3,665, $p \le 0.0001$) and TOC (F(3,24) = 98.81, MSE = 2,412, $p \le 0.0001$) 481 0.0001) removal efficiency within the pilot-scale wetland (Table 6, Figure 6). Post-hoc Tukey's-b tests 482 confirmed that significant increases in removal efficiency for each of these pollutants occurred in the 483 order NA < IA < UA < PA. Across the NA to PA aeration configurations, the mean removal efficiency 484 increased from 37 % to 92 % for COD, from 45 % to 98 % for BOD $_5$ and from 46 % to 92 % for TOC. In 485 parallel with increased pollutant removal efficiency, final effluent concentrations of COD, BOD₅ and 486 TOC decreased significantly across the NA-IA-UA-PA aeration configurations, with the lowest final 487 effluent concentration for each parameter achieved under the PA configuration (Table 6). Sample 488 position also significantly influenced pollutant removal efficiency for COD (F(2,24) = 364.47, MSE = 489 3,787, *p* ≤ 0.0001), BOD₅ (*F*(2,24) = 512.26, *MSE* = 5,197, *p* ≤ 0.0001) and TOC (*F*(2,24) = 197.77, *MSE* 490 = 4,828, $p \le 0.0001$). Post-hoc Tukey's-b tests revealed significantly higher COD, BOD₅ and TOC 491 removal efficiencies within cells one and two compared to cell three. Further, a significant 492 interaction effect between aeration configuration and sample position through the system was also 493 observed in terms of pollutant removal efficiency for COD (F(6,24) = 62.18, MSE = 645.98, $p \le$ 494 0.0001), BOD₅ (F(6,24) = 82.00, MSE = 831.95, $p \le 0.0001$) and TOC (F(6,24) = 35.55, MSE = 819.09, p495 \leq 0.0001). Post-hoc Tukey's-b tests revealed that pollutant removal efficiency was significantly 496 higher within treatment cells one and two compared to cell three for COD, BOD₅ and TOC under all 497 aeration configurations, except for the IA configuration in which no significant difference between 498 treatment cells two and three was observed. 499

500

Table 6. Summary of influent concentration, final effluent concentration and mean pollutant removal efficiency (%) for chemical oxygen demand (COD), five-day biochemical oxygen demand (BOD₅) and total organic carbon (TOC) within a pilot-scale aerated wetland operating under four different aeration configurations (n = 3 for each aeration configuration)

		COD			BOD₅			тос	
Aeration configuration	Influent (mg L⁻¹)	Final effluent (mg L ⁻¹)	Removal efficiency (%) ^(a)	Influent (mg L⁻¹)	Final effluent (mg L ⁻¹)	Removal efficiency (%) ^(a)	Influent (mg L⁻¹)	Final effluent (mg L⁻¹)	Removal efficiency (%) ^(a)
Phased (PA)	1,217 ± 28	98 ± 13	92 ± 1	834 ± 63	21 ± 5	98 ± 1	430 ± 11	35 ± 6	92 ± 2
Uniform (UA)	1,130 ± 48	246 ± 25	78 ± 1	727 ± 19	36 ± 6	95 ± 1	575 ± 106	68 ± 13	88 ± 4
Inlet-only (IA)	1,193 ± 6	676 ± 21	43 ± 2	812 ± 29	421 ± 61	48 ± 7	544 ± 57	266 ± 10	51 ± 6
None (NA)	1,161 ± 23	730 ± 44	37 ± 4	868 ± 21	477 ± 55	45 ± 6	796 ± 18	428 ± 35	46 ± 6

03 ±

(a)



± 1 standard deviation of the mean a _____ No aeration b _____ No aeration

Cumulative pollutant removal efficiency (%) determined from influent to final effluent concentration, see Equation 1





506 Figure 6. Mean reduction of (a) chemical oxygen demand (COD), (b) five day biochemical oxygen

507 demand (BOD₅) and (c) total organic carbon (TOC), concentrations (mg L⁻¹) throughout the pilot-

scale aerated wetland from influent to final effluent (cell 3 outlet, Figure 4), when tested underdifferent aeration configurations.

510

511 3.4. Impact of Hydraulic Loading Rate and Influent Strength on Pollutant Removal512 Efficiency

513

A summary of the results from the nine different optimisation tests (three hydraulic retention 514 times * three influent strengths, see Table 4) is reported in Figure 7. A significant effect of HRT on 515 516 pollutant removal efficiency was observed for COD (F(2,18) = 105.40, MSE = 1,467, $p \le 0.0001$), BOD₅ 517 $(F(2,18) = 98.40, MSE = 1,892, p \le 0.0001)$ and TOC $(F(2,18) = 28.00, MSE = 989.39, p \le 0.0001)$. Post-518 hoc Tukey's-b tests revealed significantly higher pollutant removal efficiencies within tests operating 519 with a HRT of 2.24 d and 1.49 d for all three parameters, compared to tests with a HRT of 1.14 d. No 520 significant effect of influent concentration on pollutant removal efficiency was observed for COD, 521 BOD₅ and TOC, whilst there was also no significant interaction effect between HRT and influent 522 concentration on the removal efficiency of these parameters.

523 A significant effect of HRT on the final effluent concentrations from the outlet of treatment cell 524 three was observed for COD (F(2,18) = 69.11, MSE = 565,484, $p \le 0.0001$), BOD₅ (F(2,18) = 53.50, MSE = 302,871, $p \le 0.0001$) and TOC (F(2,18) = 18.86, MSE = 79,994, $p \le 0.0001$). Post-hoc Tukey's-b tests 525 526 revealed that final effluent concentrations were significantly lower for COD, BOD₅ and TOC during 527 tests with HRTs of 2.24 d and 1.49 d, compared to tests with an HRT of 1.14 d. In contrast to 528 pollutant removal efficiency, influent concentration had a significant effect on final effluent 529 concentration for COD (F(2,18) = 21.59, MSE = 176,618, $p \le 0.0001$), $BOD_5(F(2,18) = 13.23$, MSE = 13.23, MSE = 13.274,891, $p \le 0.0001$) and TOC (F(2,18) = 7.59, MSE = 32,183, p = 0.004). Post-hoc tests revealed that 530 531 final effluent concentrations were significantly lower during tests conducted with L influent strength 532 compared to tests with M and H strength influents for both COD and TOC. Final effluent BOD₅ concentrations were significantly lower in tests conducted with L and M strength influents in 533

534	comparison to H strength influents. Despite the significant effect of HRT and influent concentration
535	on final effluent concentrations, no significant interaction effect between these factors was
536	observed in terms of the final effluent concentrations of COD, BOD₅ and TOC.
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565 (mg L⁻¹), when tested with mean BOD₅ influent concentrations of 831 mg L⁻¹, 1,355 mg L⁻¹ and 1,853

566 mg L⁻¹ representing low (L), medium (M) and high (H) influent strength and three different hydraulic
567 retention times (1.14 d, 1.49 d and 2.24 d).

568

569 4. Discussion

570

- 571 4.1. The Impact of Artificial Aeration on Pollutant Removal within Constructed572 Wetlands
- 573

574 The research reported here demonstrates the essential role of artificial aeration within HSSF 575 constructed wetlands treating high-strength organic wastewater, in order to optimise pollutant 576 removal efficiency and to achieve pollutant concentrations in final effluent that meet the typical 577 requirements of legislation. Without artificial aeration, anaerobic conditions developed throughout 578 the pilot-scale aerated wetland at Manchester Airport, indicating limited availability of DO to 579 support aerobic respiration and thereby resulting in sub-optimal removal efficiencies for COD, BOD₅ 580 and TOC (< 51 % pollutant removal compared to influent conditions). These findings are consistent 581 with the results of pilot-scale constructed wetlands dosed with de-icer contaminated runoff from 582 Edmonton Airport in Canada and Buffalo Airport in the USA, where mean BOD₅ removal efficiencies 583 of only 55 % and 68 % respectively were achieved in the absence of artificial aeration (Higgins et al., 2007). 584

However, our research also reveals that the precise configuration of artificial aeration can
significantly influence pollutant removal within HSSF constructed wetlands. Specifically, and for the
first time in an aerated wetland, we provide evidence for the advantages of a phased aeration
configuration compared to alternative aeration configurations. In contrast to the IA configuration,
both PA and UA configurations resulted in significantly higher pollutant removal efficiencies for COD,
BOD₅ and TOC. Whilst high removal efficiencies were achieved for these pollutants under the UA

591 configuration, Figure 6 emphasises that the removal of organic compounds occurs predominantly 592 within the first two-thirds of the treatment system. This is consistent with findings from previous 593 research in aerated wetlands, where up to two-thirds of organic matter was removed within the first 594 quarter of a system (Akratos and Tsihrintzis, 2007, Zhang et al., 2010). In fixed film systems, such as 595 aerated wetlands, biomass growth and microbial respiration of BOD₅ decrease exponentially 596 towards the system outlet, associated with progressive filtration of particulate organic matter and 597 declining concentrations of biodegradable organic carbon (Kadlec and Wallace, 2009). These 598 characteristics potentially result in sub-optimal operating conditions under the UA configuration, 599 involving under-aeration at the inlet zone, resulting in the development of anaerobic conditions, 600 alongside over-aeration towards the outlet of the system resulting in unnecessary aeration, energy 601 consumption and operating costs. In contrast, PA better aligns the delivery and demand for aeration 602 across a treatment system, for example delivering 50 % of the total aeration to the first third of the 603 pilot-scale system used in the research reported here. The PA approach has similarities with tapered 604 aeration designs in biological reactors, such as the activated sludge treatment process, in which 55 % 605 to 70 % of the total air input is typically applied to the first half of a system to address the high O_2 606 demand near to the inlet (Wolter and Hahn, 1995).

607 The PA configuration evaluated in our research enhanced pollutant removal efficiency by 15 %, 3 608 % and 5 % for COD, BOD₅ and TOC, compared to the results obtained under the more conventional 609 UA configuration, although pollutant removal efficiency was only statistically higher for COD under 610 the PA configuration. Despite the enhanced performance of PA compared to UA, IA or NA 611 configurations, DO concentrations within the first treatment cell remained at 0 mg L⁻¹. This indicates 612 high rates of aerobic respiration and insufficient input of air to meet the O₂ demand exerted by the 613 influent within this cell, even with PA. Further, mean DO concentrations within the media pore space 614 of cell three were high (8.3 mg L⁻¹) during PA tests, suggesting excessive inputs of air compared to 615 the O_2 demand exerted by the waste water within this cell. Therefore, opportunities remain to 616 further optimise the delivery of aeration as part of a PA configuration, in order to better match the

supply of and demand for DO through a treatment system. For example, aeration devices could be
automated to operate only when DO concentrations are within a pre-defined range, switching off
when DO concentrations are outside of this range to prevent excessive input of air to a treatment
system.

621

622 4.2. Impact of Aeration Rate and Media Depth on Standard Oxygen Transfer

623 Efficiency

624

625 Whilst our research demonstrates the important role that artificial aeration plays in optimising 626 constructed wetlands for the treatment of high-strength wastewaters, typical aeration devices 627 consume approximately 0.2 kWh of energy per m³ of water treated (Wallace et al., 2006, Murphy et 628 al., 2012) and can contribute up to 80 % of the total operating costs of a system. The optimisation of 629 design factors that control the efficiency of O_2 transfer from the gaseous to the liquid phase is 630 therefore integral to achieving low cost, sustainable treatment solutions. Our research demonstrates 631 that aeration rate was an important control on SOTE within the media-filled column experiments, in which SOTE decreased significantly from 2.4 % to 1.6 % in 1,500 mm deep columns and from 4.9 % 632 633 to 2.9 % in 3,000 mm deep columns when aeration rate increased from 1 L min⁻¹ to 3 L min⁻¹. The 634 response of SOTE to increasing aeration rate is consistent with previous results within 1,000 mm 635 deep, 2.25 m³ media-filled tanks in which SOTE decreased from 14.0 % to 5.5 % when airflow rates 636 increased from 10 L min⁻¹ to 20 L min⁻¹ (Butterworth et al., 2013). These authors also showed that 637 SOTE decreased when media diameter increased (Butterworth et al., 2013), explaining the higher 638 SOTE with the 2 mm diameter media used by these authors compared to the results reported in the 639 current paper which used 10 mm to 15 mm diameter media. The response of SOTE to increasing 640 aeration rate is also consistent with previous studies conducted within open-water diffused aeration systems, which have been examined more extensively than media-filled systems. For example, SOTE 641 decreased from 23.6 % to 18.3 % when aeration rate increased from 0.4 L min⁻¹ to 2.3 L min⁻¹ within 642

a full-scale, 2,700 mm deep oxidation ditch operating under an extended aeration configuration
(Gillot and Héduit, 2000). Further, SOTE decreased from 8.9 % to 7.1 % in a 1,500 mm deep system
and from 6.0 % to 4.5 % in a 2,900 mm system when aeration rate increased from 10 L min⁻¹ to 40 L
min⁻¹ within a 3,000 mm deep pilot-scale hypolimnetic aeration system (Ashley et al., 2008).

647 In open water systems, the aeration rate influences the fluid dynamics of air bubbles, with larger 648 bubbles being produced when the aeration rate through an orifice or diffuser is increased (Ashley et 649 al., 2008). As bubble diameter increases, the buoyancy force increases bubble terminal velocity, 650 which in turn minimises bubble retention time within a water column. Further, the bubble surface 651 area to volume ratio decreases as bubble diameter increases, resulting in a reduction in the relative 652 surface area across which the mass transfer of O_2 from the gaseous to the liquid phase can take 653 place (Gillot and Héduit, 2000, Ashley et al., 2008, Henze et al., 2008). Bubbles also form more slowly 654 when air comes into contact with water at a diffuser orifice under low airflow rates compared to 655 high airflow rates (Davidson and Schüler, 1997), presumably resulting in greater O_2 transfer during 656 the formation of each individual bubble at the diffuser orifice location (Ashley et al., 1991, Gillot and 657 Héduit, 2000). Further, more uniform distribution of air bubbles released from a diffuser orifice can 658 be achieved under low airflow rates, resulting in a more uniform distribution and greater separation 659 between individual bubbles rising through the water column. This serves to reduce bubble 660 coalescence in open-water systems (Ashley et al., 1991, Gillot and Héduit, 2000, Butterworth et al., 661 2013). However, this is likely to be less important in media-filled systems where bubble hold-up 662 within the media pore space increases bubble coalescence (Fujie et al., 1992, Collingnon, 2006, 663 Butterworth et al., 2013).

664 Generally, higher SOTEs have been reported within open water systems compared to media-filled 665 systems. However, direct comparisons between the two types of system are complicated by the very 666 different physical characteristics of open water and media-filled systems. A previous study that 667 directly compared SOTE between open water systems and media-filled systems within 1,000 mm 668 deep tanks, established that SOTE was enhanced within the media-filled system compared to the

669 open water system under aeration rates ranging from 10 L min⁻¹ to 60 L min⁻¹ (Butterworth et al., 670 2013). In media-filled systems, the media pore space can enhance bubble hold-up compared to open 671 water systems, thereby increasing bubble retention time and potential O₂ mass transfer from the 672 gaseous to the liquid phase. The effect of increased bubble retention time in the research reported by Butterworth et al appeared sufficient to negate the adverse effects of bubble coalescence on 673 674 SOTE within media-filled systems compared to open water systems. Regardless of SOTE, media-filled 675 systems offer several advantages compared to open water systems for the treatment of high-676 strength wastewater, such as enhanced sedimentation and filtration of particulate load (Faulwetter 677 et al., 2009). The presence of media also serves to provide a more robust and stable attached 678 microbial population, due to the high surface area of the media surfaces compared to open water 679 systems where microbial colonies are typically in suspension and can be more difficult to maintain 680 (Metcalf and Eddy Inc, 1991). Further, media-filled systems such as aerated wetlands typically 681 provide a higher and more consistent pollutant removal efficiency, alongside lower final effluent 682 pollutant concentrations, compared to conventional open water systems such as lagoons (ACRP, 683 2013, Freeman et al., 2015).

684 Another means of enhancing SOTE is to increase bubble retention time by increasing the depth of 685 a treatment system, thereby prolonging the time taken for a gas bubble to rise through a water or 686 media-filled column. For example, our research demonstrated that SOTE more than doubled from 687 2.42 % to 4.90 % when media depth increased from 1,500 mm to 3,000 mm at airflow rates of 1 L min⁻¹ within the media-filled columns, although we note that no statistically significant effect of 688 689 media depth on SOTE was found. Whilst only limited research has assessed the effect of media 690 depth on SOTE, the increase in SOTE with media depth reported in this paper is generally consistent 691 with the results of previous work in open water systems. For instance, SOTE increased from 4.0 % to 692 4.6 %, when diffuser depth increased from 0.24 m to 0.32 m below the media surface within a 240 693 mm internal diameter laboratory scale column operating under aeration rates of 1.6 L min⁻¹ (Zhen et 694 al., 2003). Similar findings were observed during laboratory tests conducted within 300 mm internal

diameter columns characterised by low water depths and air flow rates of 1 L min⁻¹, in which SOTE
increased from 3.9 % to 4.2 % when depth increased from 0.45 m to 0.60 m (Atta et al., 2011).

697 The substantial increase in SOTE with increased media depth that is reported here suggests that 698 more efficient, cost-effective and sustainable treatment of wastewater could be achieved through 699 increasing the media depth within artificially aerated HSSF wetlands. However, there remain 700 important practical challenges to constructing artificially aerated wetlands at depths > 1,500 mm. 701 Firstly, health and safety issues surrounding the structural stability of excavations (HSE, 2016) would 702 need to be addressed, resulting in stabilisation potentially being required during construction. 703 Secondly, an economic assessment would be required to determine the cost-effectiveness of 704 increasing media depth, given the additional costs for material excavation and disposal where the 705 excavated material cannot be reused on-site. Finally, issues with groundwater levels creating 706 hydraulic pressure beneath a treatment system, potentially damaging any impermeable liner, would 707 need to be considered. However, even relatively small increases in media depth were shown in our 708 research to generate substantial increases in SOTE, for example a 44 % increase in SOTE, from 2.4 % 709 to 3.5 % when media depth increased from 1,500 mm to 2,000 mm at airflow rates of 1 L min⁻¹. 710

711 4.3. Optimisation of Hydraulic Retention Time and Pollutant Mass Loading Rate712 within Artificially Aerated Wetlands

713

Significantly higher pollutant removal efficiencies for COD, BOD₅ and TOC were achieved under HRTs of 2.2 d and 1.5 d in comparison to an HRT of 1.1 d within the research reported here. No significant difference in pollutant removal efficiency between 2.2 d and 1.5 d HRT suggests that the optimal HRT within the treatment system evaluated here was 1.5 d. This is within the 1.2 d to 6.1 d range of HRTs reported previously for aerated wetlands treating effluents characterised by high influent ammonia or BOD₅ concentrations (Wallace et al., 2006, Wallace and Liner, 2011a, Murphy et al., 2016, Uggetti et al., 2016). Although pollutant removal efficiency was not significantly affected

by the range of influent concentrations tested, the concentrations of pollutants in the final effluent were significantly lower when the experimental system was dosed with low and medium strength influents, in contrast to high strength influents. These observations suggest that influent concentration and therefore pollutant MLR is a key factor determining the optimal operation of aerated wetland systems. Optimisation of biological wastewater treatment systems is typically achieved when steady-state MLRs are maintained, thereby facilitating the establishment of a microbial biomass that is fully acclimated to MLR of the influent. The results reported here indicate that optimal areal MLRs are 0.10 kg d⁻¹ m⁻² BOD₅, if the objective is to achieve a low final effluent concentration compliant with typical UK BOD₅ environmental permit limits. However, the treatment system evaluated here also performed at > 91% BOD₅ removal under areal MLRs of up to 0.23 kg d⁻¹ m⁻² BOD₅, although mean final effluent BOD₅ concentrations were 177 mg L⁻¹ which exceeds typical environmental permit limits and would therefore require tertiary treatment or discharge as a trade effluent. The long-term operation of aerated wetlands exceeding 0.20 kg d⁻¹ m⁻² BOD₅ is not recommended, due to the potential for microbial clogging of the media pore space, resulting in operational issues including hydraulic malfunctioning, surface flooding or reductions in pollutant removal efficiency (Nivala et al., 2012, Pedescoll et al., 2013).

746 5. Conclusion

747

Global population growth, urbanisation, industrialisation and climate change represent significant threats to the ability of freshwater ecosystems to provide critical services to human society. New forms of decentralised treatment, such as artificially aerated wetlands, represent a potentially sustainable approach for mitigating the impacts of wastewater derived from sources such as airports thereby protecting and enhancing freshwater ecosystems and the services that they provide to human society.

754 The research reported here examined how new approaches, based on artificially aerated HSSF 755 constructed wetlands, can enhance the treatment of high-strength wastewaters from sources such 756 as airports. Using a novel phased-aeration approach, we demonstrate how pollutant removal 757 efficiency per unit of aeration supplied can be significantly enhanced, potentially reducing the 758 treatment costs associated with artificially aerated HSSF constructed wetlands. Optimal operating 759 conditions for a pilot-scale system replicating an aerated HSSF constructed wetland were defined, 760 resulting in > 90% removal of COD, BOD₅ and TOC with a hydraulic residence time of 1.5 d and a mass loading rate of 0.10 kg d⁻¹ m⁻² BOD₅. Further, reduced aeration rate and increased bed media 761 depth were shown to enhance the transfer of oxygen from gaseous to liquid phases, thereby 762 763 promoting aerobic pollutant degradation processes within aerated wetlands. This research highlights 764 the potential of decentralised, aerated wetland technology to successfully treat high-strength 765 wastewaters, providing additional support for future development and application of the aerated 766 wetland technology to protect and restore freshwater ecosystems.

767

768

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