Long-term phosphorus removal in Land Treatment Systems: evaluation, experiences and opportunities

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Excessive release of phosphorus (P) from urban wastewater into water bodies is a significant environmental concern. Land treatment systems (LTS) have been used for the last 150 years as a low cost, and low energy tertiary wastewater treatment. However their P removal capacity is limited by soil adsorption capacity. Accurate P-removal lifespan prediction is needed to provide confidence in this technology and to support long-term wastewater treatment planning. This review outlines the history of LTS, describes the main P removal processes in soils and critically analyses the research methods used to date to assess long term P removal in soil. Finally, an assessment is made of the role that modelling tools can play in aiding our understanding of P processes in LTS soils and in forecasting system longevity.

KEY WORDS: phosphorus, soil, wastewater treatment, longevity, modelling

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1. INTRODUCTION

The release of excessive soluble phosphorus (P) to water bodies is a significant global environmental concern and the resultant eutrophication is one of the most common water quality problems (Smith et al. 2006; Carpenter & Bennett 2011). The consequence of eutrophication is the degradation of water resources by toxic algal blooms, excessive aquatic plant growth, oxygen depletion, death of aquatic life, and consequent loss of biodiversity. Together these water quality impacts can cause considerable loss of value of fresh water ecosystems in terms of diminished recreational value and profit reductions of the tourist sector (Carpenter & Caraco 1998) and increased water treatment costs. Pretty et al. (2003) quantified the eutrophication damage in England and Wales to be around £155 million per year.

The sources of P in surface waters vary depending on each context, and they can be classified as diffuse and point sources. The main diffuse pollution sources in the UK are natural landscapes, agricultural fields, rural and urban surfaces, waste from non-sewered populations, farm animals and, atmospheric deposition. The principal point sources are municipal and industrial effluent. The total P load to surface waters in Great Britain is estimated to be 41.6 kt/a: municipal effluent is the largest fraction with 60.7% of the P contribution, while the agricultural and industrial contributions are 28.3% and 4.6% respectively (and others 6.4%) (White and Hammond, 2002). The total phosphorus (TP) concentration in typical municipal wastewater depends on the local water consumption and it ranges from 4-25 mg/l (Henze et al. 2008). The principal sources of P in municipal wastewater in the UK are black water (66%) and grey water (34%) (White & Hammond 2002).

Since domestic wastewater is a main source of P, the EU (EU Parliament Regulation 2004) and countries such as USA, Canada or Japan, have focused their efforts during the last decades on reducing the P content in detergents (European Commission 2012). In 2010, the Detergents

46   Regulations in the UK (UK Parlament 2010) banned the commercialization of domestic

47   laundry detergent that contained more than 0.4% of inorganic phosphates (Richards et al.

48   2015). Hence, to complement the measures to control P at source, many governments are

49   setting strict pollutant discharge limits in their environmental policies. The European Waste

50   Water Directive (European Commission 1991) sets discharge limits concentrations in

51   wastewater effluents to sensitive areas at 1-2 mg/l for TP (depending on the population).

52   However, in order to meet the standards of the receiving waters, required by the Water

53   Framework Directive (European Commission 2000) it is expected that discharge P limits will

54   have to be further reduced and in some cases it may be as low as 0.1 mg/l (Parliamentary Office

55   of Science and Technology 2014).

56   Because of the need for efficient treatment, new technologies have been developed to enhance

57   P removal from sewage but with the increased effectiveness their cost has risen. Thus, to meet

58   these legal requirements, the wastewater industry faces the problem of providing efficient but

59   low cost, low energy and low carbon wastewater treatments. Land treatment - defined by Crites

60   et al., (2000, p. 7) as the "controlled application of water onto the land surface to achieve a

61   specified level of treatment through natural, physical, chemical, and biological processes

62   within the plant-soil-water matrix"- can help to overcome these challenges, however such

63   systems have a finite capacity for P removal because of the limited adsorption capacity of the

64   soil.

65   The aim of this paper is to critically review the existing knowledge and tools available for

66   making reliable predictions about how long a Land Treatment System (LTS) can effectively

67   perform as a tertiary treatment for wastewater without compromising the quality of the nearby

68   environment and to identify the current knowledge gaps in long-term P removal behaviour of

69   these systems. In order to analyse the previous studies about long term performance of LTS,

70   the state-of-the-art is examined using the following approach: description of LTS and problems

71 associated with P forms in this type of natural wastewater treatment; analysis of current

72 knowledge regarding P removal and longevity in laboratory-scale and full-scale systems; and

73 finally, evaluation of the gaps for further research needs.

74   **2.   LAND TREATMENT SYSTEMS**

75 Since humans established permanently in communities, LTS have been used as a way to

76 manage wastewater (Inc. Metcalf & Eddy et al. 2003). Historical reports illustrate the use of

77 LTS in early Greek and Chinese civilizations as an irrigation and sanitation method

78 (Tzanakakis et al. 2007), but it was not until the second half of the 19th century when "sewage

79 farming" (a historical term used for LTS) was used to protect public health after the cholera

80 epidemics in England. This expansion drove the development of the basic principles of

81 planning, operation and management of the systems. By that time, the first LTS appeared in

82 the USA, France and Germany contributing to the development of the technology. However,

83 their use declined as a result of the development of more intensive treatments to cope with

84 population growth and lack of land availability (Jewell & Seabrook 1979). In the USA, the

85 Clean Water Act (The 92nd United States Congress 1972) renewed interest in land treatments

86 when planners realized that these systems were able to meet new discharge requirements of the

87 National Pollutant Discharge Elimination System. Recently, interest in these systems has been

88 renewed due to the possibilities that they offer as on-site treatments for remote areas and small

89 communities, overcoming the high energy cost of conventional systems. In addition, they have

90 been used as a tertiary treatment for polishing effluent from Wastewater Treatment Plants

91 (WWTP), in some cases utilising the supply of nutrient-rich water for biomass production for

92 economic benefit (Paranychianakis et al. 2006; Nissim et al. 2015).

93 During the period 1920-1970, LTS were used as a tertiary treatment in the UK to provide

94 confidence with respect to meeting the so-called Royal Commission 20:30 biological oxygen

95   demand: suspended solids discharge consent standard which was widely applied at British

96   wastewater treatment works (Gray 1989). Many of these LTS were subsequently inherited by

97   the regional water authorities following the Water Act (1973) and remain today. Sweaney

98   (2011) gathered data in relation to organic matter, suspended solids and ammonia removal from

99   20 LTS used for tertiary treatment by UK water companies and found that the systems did not

100   have any formal design and used different applications rates and sizes (probably due to the lack

101   of official guidance), this made it difficult to draw conclusions about any performance trend.

102   The study however did not show results for P (Sweaney, 2011).

103   Compared to mechanical or chemical wastewater treatments, LTS are cheaper to operate and

104   maintain, whilst also being less energy demanding (Sapkota et al. 2014; Tzanakakis et al.

105   2007). Their main constraint is that they require larger amounts of land than conventional

106   centralised wastewater treatments, so they have largely been used in rural areas, for single

107   household or small-scale applications, where land availability is not a problem. They have been

108   successfully implemented for small communities (< 10 000 p.e.) in Scandinavia, the UK,

109   Australia and USA.

110   Depending on the loading rate and flow path, LTS can be classified, into: slow rate (SR),

111   overland flow (OF) and rapid infiltration (RI) systems (Smith 2006). Each achieves different

112   performance outcomes and can/not be implemented, depending on the site characteristics and

113   the desired level of treatment (Table1). In LTS, the main treatment mechanisms are: chemical

114   retention and transformation, mechanical retention and biological transformation. While

115   mechanical retention (e.g. grass filters strips) and biological transformation can treat pollutants

116   such as solids, organic matter (OM), nitrogen and pathogenic microorganisms; adsorption and

117   precipitation are the main processes responsible for P removal in soils (Vohla et al. 2011).

118   These mechanisms differ from soil to soil and depend significantly on soil surface chemistry

119   and pH.

120   Each type of soil is characterized by a P fixation capacity and is dependent on the number of

121   reactive sites that can react with the phosphate. It depends on the amount and type of clay

122   present, the soil pH and the OM content. When all the P-fixation sites are filled, the maximum

123   phosphorus-fixing capacity of the soil is reached, thus the soil is supersaturated with P (Brady

124   & Weil 2008). Consequently, P removal in LTS is a finite process (Drizo et al. 2002). In these

125   situations the required level of P removal is not achieved and excess P can be released to the

126   environment either in leachate or runoff as dissolved P.

127   Although longevity of the LTS is one of the key parameters to achieve cost-effective

128   treatments, little research has been carried out regarding effects of long term wastewater

129   application in LTS and the longevity of the system in terms of P removal (Hu et al., 2006).

130   Three key questions regarding long-term P removal in LTS remain (Hu et al. 2005; Hu et al.

131   2006; Robertson 2012; Weiss et al. 2008; Moura et al. 2011; Eveborn et al. 2012; Eveborn et

132   al. 2014): i) is the required knowledge available about how long one of these systems can

133   immobilize P?, ii) what are the mechanisms involved and what is their relative importance?

134   and iii) what should be the approach to estimate of the P removal capacity by LTS in the long

135   term?

136   **3. PHOSPHORUS REMOVAL PROCESSES IN LAND TREATMENT SYSTEMS**

137   The main removal phosphorus mechanisms in natural treatments are vegetation uptake,

138   microbial processes, precipitation and adsorption (Reddy et al. 1999). In overland flow systems

139   vegetation uptake removal pathway can be significant (~20-30%) if vegetation is harvested

140   regularly (Crites et al. 2006). Precipitation is the process in which phosphates can be removed

141   from the soil solution when it reacts with Fe, Al, Mn (acidic soils) or Ca (basic soils) to form

142   phosphate minerals. This process is considered to be irreversible but depends on pH, redox

143   conditions and the concentration of the mineral ions in solutions required to precipitate

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144   (McCray et al. 2009). Adsorption is the process of fixation (chemisorption) of PO_4 ions to soil

145   particles. P interacts with soil particles in its exchangeable orthophosphate form, the pH drives

146   the availability of each orthophosphoric form in the soil solution H_3PO_4, $H_2PO_4^{-1}$, HPO_4^{-2} and

147   PO_4^{-3}, at soil pH, PO_4^{-3} has the strongest binding capacity (MacBride 1994). Positive charged

148   soils with Fe, Al, Ca content attract phosphate (PO_4) anions but they can also be adsorbed to

149   the soil surface through non-electrostatic forces (Evangelou 1998). Adsorption is fast process

150   but desorption is usually slower. It is driven by an equilibrium constant and if the concentration

151   of PO_4 ions rise in the soil solution, the ion will adsorb to soil charged surfaces to restore the

152   equilibrium. Adsorption is limited by the number of available sorption sites (McCray et al.

153   2009)

154   Despite the numerous studies and significant knowledge regarding all these processes, it is

155   complicated to predict P removal performance in LTS as different environments (soil pH, soil

156   type, temperature, etc.) will promote different processes that operate at different rates (McCray

157   et al. 2009). Current understanding of soil P availability to plants gives us a conceptual model

158   to help us to understand P dynamics in LTS. Syers, Johnston & Curtin, (2008) proposed that

159   phosphorus in soil can be considered to be in one of four different pools on the basis of its

160   availability to the plant: immediately available P in soil solution, readily available P that is

161   adsorbed weakly at the soil surface, more strongly bound P that is less available, and finally

162   very strongly bound P that is either of very low availability or inaccessible. High concentrations

163   of P in soil solution due to wastewater irrigation (~10 mg P/l) will encourage an initial rapid

164   phase of P sorption on high-affinity sites on the soil surface (Paranychianakis et al. 2006). The

165   positive charges associated with soils with a high anion exchange capacity allow rapid, readily-

166   reversible and non-specific electrostatic adsorption of anions such as phosphate. These sorption

167   processes are pH dependent and are likely to be associated with the surfaces of Fe and Al

168   minerals in strongly acid to neutral systems and on Ca minerals in neutral to alkaline systems

169   (Lindsay 1980). A slower phase of P removal is attributed to phosphorus diffusion in poorly

170   accessible sites and/or to chemical precipitation which apportions P to the strongly-bound pool

171   (Paranychianakis et al. 2006; Syers et al. 2008). Inorganic P compounds in soils are commonly

172   associated with iron, aluminium, and calcium. These compounds have a high degree of

173   variability in their solubility and stability which are influenced by pH. Those compounds with

174   very low solubility, such as apatite $(Ca_{10}(PO_4)_6(OH,F,Cl)_2)$, would be associated with the very

175   low availability pool.

176   One of the key issues related to the study of lifespan of LTS is the time dependency component

177   in the P immobilization reaction (Paranychianakis et al. 2006; Cucarella & Renman 2009). It

178   consists of a two-step reaction: an initial and fast sorption phase and a slower phase related to

179   P diffusion or/and chemical precipitation. However, the secondary slow or irreversible sorption

180   that has been widely documented in these laboratory studies was not evident in long term P

181   monitoring in septic tanks plumes in Canada by Robertson (2008). Consequently, Robertson

182   concluded that, secondary P attenuation processes, such as P diffusion into soil particle

183   microsites, slow recrystallization of sorbed P to insoluble metal phosphate minerals, or slow

184   direct precipitation could be inactive in ground water zones or too slow to be observed in the

185   16-year period of the study, suggesting that P can be remain mobile for decades with the

186   consequent risk of impact to nearby rivers and lakes.

187   If soil characteristics change over time, P sorption capacity (PSC) may not remain constant,

188   which has implications for LTS lifetime estimations. Changes in physical and chemical soil

189   properties due to continuous application of wastewater have been characterized from previous

190   studies. These changes may also affect the P sorption capacity of the soil which is often

191   assumed to remain constant for a certain site during the lifespan of the LTS (Hu et al. 2005).

192   Hu et al. (2006) described an increase in pH from 5.6 to 7.0 in the top 15 cm layer of the soil

193   after more than 30 years of high pH (~8.5) wastewater application coming from a paper mill

194 (25%), other industries (~25%) and (~25%) from domestic wastewater in the Muskegon

195 Wastewater Treatment facility (USA) with major soil types categorized as sands. These results

196 are consistent with those of Richardson et al. (1988) who reported statistically significant

197 increases ($P>0.95$) in North Carolina in three acidic mineral soils at a coastal plain swamp after

198 municipal wastewater additions for 30 years. On the contrary, Eveborn et al. (2014) in their

199 study of P accumulation and mobility in soil treatment systems after 8 to 11 years of domestic

200 wastewater application in sand filter beds, revealed that the pH of the top layer was between 1-

201 2 units lower than in reference samples. Their interpretation in this case was that the pH change

202 was due to the chemical dissolution and consumption of reactive calcium oxide and other

203 alkaline minerals or acidifying organic degradation processses. However, the study points out

204 that the results in the surface layer might be also have been affected by surface regeneration

205 works carried out during the study. Moreover, due to long-term high salt content effluent

206 application in the Muskegon studies, exchangeable Ca concentrations increased ten fold

207 (Zhang et al. 2007; Hu et al. 2005; Hu et al. 2006). Hu et al. (2006) suggests that the increase

208 in the pH might have as a consequence the increase in the exchangable Ca in the topsoil and

209 links this with the possibility of an extension of the lifespan of the LTS. The experiments of

210 Eveborn et al. (2014) also suggest that the increase in oxalate extractable Al and Fe could be

211 due to weathering mechanisms provoked by acidification of the soil after the wastewater

212 additions, which will also affect the sorption capacity of the soil.

213 It is clear that efficiency of P removal declines with time, with slower sorption processes

214 approaching the equilibrium, and that a number of factors affect it (soil mineral type, amount

215 of clay, pH, OM, anion presence or temperature). However, Drizo et al. 2002 and Hu et al.

216 2005 pointed out the possibility of long-term P accumulation and removal capacity

217 regeneration. The proposed mechanism depends upon the formation of new P-retentive surface

218 clusters through phosphate compound adsorption that act as new reactive sites for adsorption

219　and precipitation. Eveborn et al. 2014 also suggests that P bound to the surface in stable pools

220　may in the long term increase the P sorption capacity of soils due to changes in pH and in

221　oxalate-extractable metals during the wastewater application. Furthermore, Drizo et al. 2002

222　demonstrated that a wetland constructed using electric arc furnace slag as a filter medium got

223　back 74% of its P retention capacity after four weeks rest and claim that other studies observed

224　that soils had restored their P adsortion capacity after repeated wetting and drying cycles. The

225　mechanisms involved are not well understood, but it is suggested that it is related with elevated

226　pH after drainning the material that will bring Ca, Fe and other mineral ions in supersaturation

227　with the solution to creat new sites for P adsorption/precipitation in the filter pores Bisone et

228　al. (2016) studied behaviour of a clay soil with wastewater additions and the influence of

229　hydraulic loading. The study focused on the evaluation of wetting and drying cycles in clay

230　soil used for wastewater treatment and results indicated the potential to adsorp P and that

231　saturation and desaturation cycles did not influence phosphate retention but enhaced

232　nitrification. However, the soil saturation can lead into P desorption caused by Fe reduction

233　and therefore intermittent loading can favour infiltration and help to avoid desorption

234　Therefore, LTS design and management approaches which permit resting periods could allow

235　the use of clay soils as a longer term polishing treatment for P removal.

236　**4. METHODS TO ASSESS PHOSPHORUS REMOVAL CAPACITY IN SOILS**

237　The standard tests to assess P transport in soils (such as water extractable phosphate,

238　exchangeable phosphate, sorption and desorption isotherms) are usually based on agronomic

239　aspects of P availability for the plant and not to determine environmental risk of P loss in soils

240　(Moura et al. 2011). Although they can be used to quantify the nature and quantity of P

241　concentrations in the soil, a better understanding is needed to determine which of those indices

242　are more suitable to better assess long-term P fate in LTS.

243   Hooda et al. (2000) studied the potential of different soil tests to predict P release to water of

244   different soils in the UK that had received long-term fertilization or sewage sludge: Soil Test

245   P (STP), sorption–desorption indices, the degree of saturation of P (DPS) and the amount of

246   water dissolvable-P. The study discussed that the DSP of the soils is more important than other

247   indices because soil extractable-P does not integrate the P soil characteristics, but the DSP

248   index associates P management and soil type factors helping to identify soils that are likely to

249   become diffuse sources. Moura et al. (2011) studied the effects of long-term reclaimed water

250   application on P leaching potential in rapid infiltration basins, the study concluded that

251   extractable P and P-saturation ratio are good indicators of soluble concentrations in the

252   leachates. Furthermore, Hu et al. (2005) reported that labile P distribution is not a good

253   indicator of P leaching in soil wastewater treatments since it is not correlated with TP, Fe-

254   bound P, or Ca/Mg-bound P, but, identified the need for a new index that integrates DSP and

255   Ca adsorption.

256   Traditionally, the P removal capacity of the soils has been related to the absorption-adsorption

257   P capacity. This can be determined by batch-scale experiments where the soil interacts with

258   solutions at different P concentrations until the equilibrium is reached, and translated to

259   Langmuir or Freundlich equations to represent their kinetic relationship (Kovar & Pierzynski

260   2009). However, many researchers agree that there is a lack of studies advising how to interpret

261   and use such batch experimental results to assess long term effects and life expectancy of soil

262   infiltration systems (Zhang et al. 2007; Cucarella & Renman 2009; Hu et al. 2006; Eveborn et

263   al. 2012; Drizo et al. 2002).

264   Nair et al. (1984) identified a wide range of methodologies that had been used to determine P

265   adsorption. Although diverse methodologies made the comparison among studies difficult,

266   they proposed a standard method to predict the partitioning of the dissolved inorganic P. In

267   addition, Graetz & Nair (2000) pointed out that the laboratory procedures to determine the P

268   adsorption had advantages; such as the possibility to separate the soil and solution or the

269   possibility to obtain the necessary volume sample for carrying out the tests. However, they also

270   described experimental disadvantages such as particle breakdown while shaking. The study of

271   Hooda et al. (2000) asserts that phosphorus adsorption-sorption isotherms are useful to

272   compare soil characteristics but their empirical nature make them unsuitable to explain

273   mechanisms of sorption-desorption of P in soils as they do not reflect field conditions like

274   runoff or rainfall.

275   The Onsite Wastewater Treatment Systems Manual from the Office of Research and

276   Development of the U.S. Environmental Protection Agency (2002) states that the estimation

277   of soil capacity removal can be based on sorption isotherms but it totally underestimates the

278   capacity of P removal since they do not reflect the slow reactions that take place (the standard

279   procedure extraction time is 24h) and declare that studies revealed that, in the long term, the

280   capacity could be extended from standard isotherms predictions by 1.5-3 times. Similarly, Hu

281   et al. (2006) evaluated the effects of long-term wastewater application on sorption capacity of

282   the soil by comparing the 1-day maximum sorption capacity (P_{max}) before and after long-term

283   wastewater application. The study revealed that the 1-day P_{max} increased by ~3 times since the

284   start of wastewater application indicating that it is possible for the soil to continuously adsorb

285   P and therefore, difficult to get a reliable estimate of P_{max}. Ádám et al. (2005) attempted to

286   estimate the long-term P sorption capacity of filter materials. The study criticizes the use of 24-

287   h sorption batch tests claiming that results differ from full-scale systems. To get more reliable

288   long-term P sorption estimations they proposed a long-term experiment of a full scale

289   horizontal constructed wetland (CW) and also concluded that batch experiments are not

290   appropriate to measure sorption capacity materials because the experimental parameters (e.g.

291   soil solution P concentration and water:soil ratio) are usually higher than in full-scale systems.

292   However with full-scale constructed wetlands the extrapolation of the results are more difficult

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293 because of the complexity of the systems due to biofilm developments or the presence of plants

294 that can enhance the P removal but can also generate cracks or pores in the material leading to

295 preferential flow (Ádám & Krogstad 2006).

296 Cucarella and Renman (2009) exhaustively and critically reviewed batch experiments used to

297 estimate the PSC of filter materials used in on-site wastewater treatments. The study confirmed

298 the findings of Nair et al. (1984) by certifying that different studies are not comparable because

299 they are arbitrarily run under different experimental conditions and with different experimental

300 parameters such as, solution-ratio during shaking, P concentrations of the soil water, contact

301 time, agitation characteristics and temperature. As those parameters have the potential to

302 significantly influence P processes they should be properly established in the experimental set

303 up to reduce their influence in the PSC determination. Additionally, experiments should, as

304 much as possible, represent real field conditions. Drizo et al. (2002) recommended to couple

305 batch experiments with long-term column experiments for P removal efficiency studies in order

306 to integrate the effect of soil chemical and hydrological properties.

307 Column experiments are frequently used to determine hydrological properties, evaluate

308 transport models and monitor fate and mobility of pollutants in soil (Lewis & Sjöstrom 2010).

309 However, Zhang et al. (2007) criticised previous column tests used to predict the fate of P in

310 slow rate wastewater treatment soils. The tests used high hydraulic loading rates (up to 250

311 times those typically used slow rate systems) and were fed continuously, which does not

312 correspond with typical LTS management parameters and, according to Hu et al. (2006), these

313 column tests were not run for long enough to allow slow P precipitation. Eveborn et al. (2014)

314 set up experimental columns to study the P removal and leaching potential of soil materials

315 using deionized water. Results indicated that P removal in unsaturated soil depends on P

316 loading, and that wash-out processes can take place in these systems, for example, during rain

317 events and that dissolution of aluminium phosphates and the shift in ionic strength could be a

318   possible P release mechanism. Drizo et al. (2002) aimed to determine the long-term P saturation

319   of filter materials through column experiments. The study revealed the advantages of P

320   saturation obtained from column experiments compared to batch experiments, for example

321   particles do not breakdown as a result of the shaking and the saturation is progressive. The

322   study finally recommended P feeding concentration in column tests between 40-400 mg/l to

323   reach the saturation point of filter materials in a practical timescale and experiments lasting 3-

324   5 months to allow time for slower P retention processes to occur.

325   Direct mass balance calculations ($P_{accumulated} = P_{inlet} - P_{outlet}$) of long-term performance systems

326   have also been used to assess the P removal capacity of soils although few attempts of

327   evaluating the P removal performance of full-scale long-term sewage effluent irrigated soils

328   have been reported. Kardos & Hook (1976) quantified the P added, removed by crops or

329   leached over 10 years of irrigation with treated municipal wastewater. They concluded that

330   soluble P concentrations at 120 cm depth in a clay loam soil remained close to 0.05 mg/l and,

331   <2% of the P added was leached in 10 years without vegetation removal. The same experience

332   in sandy loam reported 3% leaching. Hu et al. (2006) performed a 27 years mass balance of the

333   Muskegon wastewater treatment plant (USA), reporting 99% retention of the P applied.

334   Eveborn et al. (2012) studied the performance of four Swedish uncovered soil treatment

335   community-scale systems aged from 14 to 22 years old. Using a mass balance approach, the

336   treatments presented much poorer P removal (8-16%) than previous soil treatment reports.

337   Robertson (2012) quantified the P retained in a lakeshore non-calcareous sand filter with water

338   table fluctuations, and compared it with the estimated lifetime after 20 years of operation.

339   Contrary to Eveborn's findings, the results showed that the filter bed had retained almost all of

340   the P loaded with the wastewater over the two decades of operation. More recently, Dzakpasu

341   et al. (2015) studied the long-term capacity and efficiency of a soil substrate full-scale

342   integrated constructed wetland in Ireland, by studying the wastewater inflow, outflow and

343   storage of P in plants and sediments. Results revealed a 91% a total P removal, where 58% of

344   P storage is accounted for P removal in the soil. One of the largest experiences with soil

345   infiltration systems is situated in Bardu (Norway), where municipal wastewater from 5000

346   inhabitants has been pumped into glaciofluvial sand and gravel deposit infiltration basins at a

347   mean temperature of +0.7ºC. After 25 years of operation the treatment performance was 99%

348   P removal and still it was estimated to last 12 years more (Jenssen et al. 2014). Reddy et al.

349   (1999) identified the main difficulties when trying to assess P retention in wetlands as: failure

350   to identify and measure inputs and outputs of P and water, use of only one form of P,

351   comparison of synchronous flows and grabbing samples from strong variable flow or

352   concentration. Therefore, the few attempts at quantifying the P removal efficiency of long-term

353   full-scale experiences in natural treatments have been reported, mainly in grasslands and

354   temperate climates, with variable results. The results comparison among studies indicates high

355   removal variability probably due to differences in soil chemical properties, age of the system

356   or sampling and calculation methodologies.

357   **5.   TOWARDS ESTIMATES OF LONGEVITY**

358   There is not a clear definition regarding life expectancy or longevity of LTS (Yu 2012). In fact,

359   there is a common misunderstanding between exhaustion of P, retention capacity of the

360   material and longevity of the system. From the point of view of on-site treatments, longevity

361   must be interpreted as the time when the system reaches the effluent P discharge limit (Heistad

362   et al., 2006), which differs among countries and legislations in terms of both concentration and

363   exceedance statistics. A number of studies have calculated longevity from a simple approach,

364   the P_{max} sorption capacity of the soil (g of P/kg), the volume of land being used for treatment

365   (m^3), P content of applied wastewater (g of P/PE) and the discharge rate (g P/day) (Table 2).

366   However, the limitations discussed in previous sections highlighted how experimental

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367 conditions in P_{max} assessment can contribute to errors in such a lifetime forecast. Cucarella &

368 Renman (2009), Vohla et al. (2011) and Chang et al. (2014) noted these limitations and

369 suggested that life expectancy calculations could not be entirely addressed through filter

370 materials characteristics and that, to forecast precisely the lifespan of the system, other factors

371 (including hydrological conditions, temperature and microbiological transformations) must be

372 taken into consideration.

373 Simulation models can help to better understand the importance of bio-physico-chemical and

374 hydraulic processes relevant to pollutant removal, while helping to further understand how

375 design parameters affect the performance of the system and predict longevity and exhaustion

376 of the material (Beach & McCray 2003). Recently, researchers have gone beyond the black-

377 box, empirical models, to develop mechanistic models that consider the hydrodynamic and

378 biodegradation process in natural treatments (García et al. 2010). They cover a wide range of

379 biokinetic, process-dedicated, and design support models (Meyer et al. 2015). Process

380 dedicated models represent degradation and transfer of compounds; hence they can help with

381 understanding the fate of P and removal in LTS. These models describe P transport and sorption

382 by soil or filter materials under different hydrological conditions and some of them predict

383 breakthrough curves (Langergraber & Šimůnek 2012; Claveau-Mallet et al. 2014; Sinclair,

384 Jamieson, Gordon, et al. 2014; Sinclair, Jamieson, Madani, et al. 2014; Liolios et al. 2015).

385 Intensive research has been produced since the late-1970's to develop models able to simulate

386 the interaction in between the soil matrix, the water transport and P. The main aims of these

387 models was to evaluate non-point source pollution of subsurface waters by agricultural P

388 (Vadas et al. 2013). Many of these models have been updated through time and continuously

389 developed. Different models have been used to accomplish the aims depending on spatial scale

390 and resolution (watershed, field-scale), time scale (long-term, short–term), and the

391 hydrological system (saturated flow, variable saturated flow, horizontal flow and, vertical

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392 flow). These models can be divided into ones that use a water balance model and the ones that

393 use numerical solution of the Richard's equation. The models based on Richard's equation are

394 more input intensive and computationally demanding than the water balance models but they

395 provide more accurate solutions for field scale modelling. Regarding the overview of soil

396 phosphorus dynamics of those models. McGechan & Lewis (2002b) provided an exhaustive

397 review of a number of field scale P dynamics models and the equations used (e.g. ANIMO

398 (Groenendijk & Kroes 1999), GLEAMS (Leonard et al. 1986), MACRO (Jarvis & Larsbo

399 2012) and DAYCENT (Parton et al. 1998)), pointing out MACRO as the most complete model

400 for soil transport processes but with more simple representations of P transformation.

401 Some of the most recent experiences of numerical modelling of on-site wastewater treatments

402 and phosphorus fate predictions are related with the use of HYDRUS (J. Šimůnek, M. Šejna,

403 H. Saito, M. Sakai 2013). HYDRUS-1D and 2D are able to simulate one and two-dimensional

404 movement of water, heat and multiple solutes in variably-saturated media and can be coupled

405 with different specific modules like wetland module (CW2D) to be able to run P degradation

406 along with other processes (Langergraber & Šimůnek 2012). Recently, Langergraber (2016)

407 reviewed process-based models applicable to subsurface flow wetlands treatment, and

408 identified the HYDRUS Wetland Module as one of the most advanced ones because of its

409 multi-component biokinetics-models and stated that it is powerful tool for understanding

410 wetland processes. PHREEQC (Parkhurst & Appelo 1999) can also be coupled with HYDRUS

411 and be used to solve complex geochemical calculations (Claveau-Mallet et al. 2014). In

412 addition, MODFLOW (Twarakavi et al. 2008) is a 3D finite-difference groundwater model that

413 can be also coupled and adapted with HYDRUS.

414 The HYDRUS-1D and 2D numerical code since version 4.0, released in 2007, is capable of

415 simulating a large quantity of non-equilibrium flow and transport processes. They can be

416 divided in physical non-equilibrium and chemical non equilibrium processes. The study by

417   Šimůnek & van Genuchten (2008) gave an overview of the conceptual models, and the

418   specifications of each model with the water flow equations, the solute transport equations and

419   the number of parameters needed in each. This new feature provides a wide range of

420   possibilities to simulate phosphorus transport in soils, such as dual permeability models to

421   simulate water flow in two different domains, with different flow velocities and non-

422   equilibrium chemical models. These models range from the simple one kinetic site model

423   described as a first-order rate equation to two sites models with different sorption kinetics for

424   different fractions, where one fraction is assumed to be instantaneous and the other one is

425   kinetic. Vogel et al. (2015) successfully simulated flow and transport in a two-dimensional

426   dual–permeability system with spatially variable hydraulic properties permitting to compare

427   different spatial distributions of hydraulic properties with model responses comparison.

428   Regarding P studies with HYDRUS Naseri et al (2011) compared physical modelling of silty

429   clay-loam texture columns against the numerical modelling of HYDRUS-3D, the results

430   concluded that it simulated the water flow in the columns successfully, but overestimated the

431   final sorbed PO_4 concentrations in the soil. Elmi et al. (2012) found similar over-estimation

432   results when using HYDRUS-1D to study the vertical distribution and transport processes of

433   PO_4 in soil columns. The simulation indicated that 98% of the P applied was accumulated in

434   the first 0.2 m of the column and decreased with depth. Sinclair et al. (2014a) also used

435   HYDRUS-2D to simulate different lateral flow sand filters in on-site wastewater treatments

436   after septic tanks showing poor removal rates and effluent exceeding 1 mg P/l concentration at

437   the end of the study. Morrissey et al. (2015) studied with HYDRUS-2D the impact of

438   unsewered cluster housing simulating the attenuation of contaminants through the unsaturated

439   zone revealing limited impact to groundwater quality, and used the calibrated model to simulate

440   the impact of increasing the house density with new developments. All those studies were

441 focused on studying transport and contamination effects but none of them introduced the time

442 variable in the study in order to predict remaining lifetime.

443 Although a number of studies point out the potential for exhaustion of the P sorption capacity

444 in treatment systems, there have only been a few attempts to forecast P longevity in soil

445 infiltration systems. The first one is related with the Muskegon (USA) (Zhang & Dahab 2006)

446 wastewater treatment plant which was part of the large-scale demonstration project for nutrient

447 removal. The project aimed to evaluate long-term P performance, as it is known to be one of

448 the most important lifespan limiting factors. The first phase of the study calculated new 1-day

449 P_{max} after 30 years of operation and the new remaining life expectancy (RLE). The new

450 estimations had an extension in the RLE because of the changes in soil properties after long-

451 term wastewater application in the soil (Hu et al. 2006). The second phase of the same study

452 consisted of column breakthrough modelling, where they re-calculated the RLE (Zhang et al.

453 2007). As a result, the study proposed a remaining life expectancy prediction formula:

454
$$RLE = t_b * F / 180 \qquad\qquad (1)$$

455 where

456     $t_{b=}$ the breakthrough day

457   180=the irrigation days per year at the Muskegon plant

458   F= the correction factor for the inlet P concentration and the concentration used in the

459   study.

460 The same project proposed a model based on the mass balance principle. The empirical model

461 is able to predict the life expectancy on a yearly basis through the effects of the main

462 operational parameters of the system (rotating crop type, hydraulic loading rate, annual

463 precipitation, soil type, etc.). However, the interaction between the soil and the hydraulic

464 systems are not reflected upon. Parkhurst et al. (2003) described a reactive-transport model

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465 developed to simulate P in the sewage plume from treated sewage effluent to ground-

466 infiltration disposal beds at the Massachusetts Military Reservation on western Cape Cod

467 (USA). The simulations covered the discharged period (1936-1995) and the 60 following years

468 after cessation of disposal. The three-dimensional reactive-transport model used to develop the

469 base-case was PHAST (Parkhurst & Kipp 2002), model limitations were associated with

470 loading, flow systems and sorption characteristics of the aquifer uncertainties. Regardless that

471 the aim was not to study the longevity of the system because the activity of the filter beds had

472 stopped, it is a useful exercise of long-term plume migration modeling. Yu (2012) attempted a

473 one-dimensional model that aimed to provide the longevity of any point of the infiltration

474 system considering a homogeneous and isotropic soil, and constant and unsaturated flow. It

475 was aimed to estimate the longevity, the exhaustion time and sorbed P. The model proposed

476 was a contaminant solute transport model where advection, diffusion, dispersion and sorption

477 reactions were represented. The simulation indicated that the most sensitive factors for P

478 concentration in a soil column to reach critical concentration were the average pore flow

479 velocity and the P sorption capacity of the soil. The longevity estimations from the model

480 compared to the service time of the infiltration study sites were shorter Herrmann et al. (2013)

481 modelled the transport and removal of PO_4 in a reactive filter using the hydro-geochemical

482 transport code PHREEQC (Parkhurst & Appelo 2011), the study claims that in order to be used

483 to forecast performance and longevity it needs to be further tested and develop due to that it

484 does not take into consideration adsorption processes or the effect of biofilms growth in the

485 filter material. Claveau-Mallet et al. (2014) proposed a prediction tool for slag filter longevity

486 based on dissolution and precipitation kinetics. The proposed methodology was based on three

487 steps: (1) experimental batch tests methodology, (2) numerical calibration of the batch test (3)

488 numerical simulations of the filter. However the study assumed several limitations and

489 highlighting the principal one as being the assumption that the contact between the water and

490 the filter particles is limited, and not being affected by clogging from precipitates or particulate

491 OM. The model needs to be calibrated before being used as a design tool and proposes to couple

492 a hydrodynamic and geochemical code such as HYDRUS-PHREEQC to overcome those

493 drawbacks.

6. SYNTHESIS AND CONCLUSIONS

495 LTS have been in use since wastewater treatments were applied to protect public health and

496 still they have a role to play at the current wastewater industry as a low energy and low

497 maintenance polishing step for small wastewater treatments. They are normally used before the

498 wastewater treatment plant effluent is released to the environment to avoid environmental

499 problems, mainly eutrophication related with excessive P concentrations.

500 P removal mechanisms in LTS are physical retention, sorption processes and microbial and

501 plant uptake. Sorption is a finite process and depends on the interaction with the environment

502 and the chemical properties of the soil (ion presence, pH, OM, clay content, etc). Thus, if this

503 conditions change over time, mainly due to long-term wastewater irrigation, the P removal

504 potential will also change.

505 It is well accepted that estimation of the P sorption capacity of soils by batch experiments has

506 several limitations when it is used to calculate the P_{max} in the long term or when used to forecast

507 the longevity of the system. The standard methodology is a short-term sorption analysis (24h)

508 that underestimates the long-term removal capacity because it does not take into account the

509 slow precipitation processes and because the experimental set up does not represent the real

510 systems and interactions with the ecosystem. Thus, studies advise that these methods are not

511 used for lifespan prediction but to compare materials and soils characteristics. Column

512 experiments should be also correctly design when addressing long-term P removal

513   characterization and filter material saturation, mainly regarding length of the experiment and

514   hydraulic loading in order to correctly translate results to full-scale experiences.

515   Regarding lifetime predictions in LTS, usually, direct mass balance approach using the P_{max}

516   has been used, however the drawbacks previously pointed out also affect this methodology.

517   Using modelling tools to assess the water flow, contaminant transformation and contaminant-

518   transport processes can help to assist in the integration of all the parameters that affect the

519   functioning of the system. However, few attempts to estimate the P longevity of LTS through

520   models have been found and none of them has been completed and published due to several

521   limitations and further development and research needed (uncertainties in data acquisition,

522   empirical models, models not validated or neglecting important variables of the removal

523   processes).

524   LTS's modelling challenge lies in getting useful results from batch and column experiment

525   regarding soil P adsorption behaviour in the long term, and how to transfer those experimental

526   results into the solute transport equation. The longevity of the LTS is one of the key parameters

527   to achieve cost-effective treatments and if new studies are able to successfully model treatment

528   scenarios then we will become a useful LTS management tool and provide greater confidence

529   in the sustainable operation of these systems required by both operators and regulators.

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7. REFERENCES

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777 **Table 1** Comparison of Land Treatment Process Design Features. **Source:** (Smith 2006)

Feature	Slow rate (SR)	Overland flow (OF)	Rapid Infiltration (RI)
Minimum pre-treatment	Primary sedimentation	Screening	Primary sedimentation
Annual loading rate (m/yr)	0.5-6	3-20	6-125
Typical annual loading rate (m/yr)	1.5	10	30
Field area required (ha)	23-280	3.6-44	3-23
Typical weekly loading rate (cm/week)	1.9-6.5	6-40	10-240
Disposition of applied wastewater	Evaporation and percolation	Evapotranspiration surface run off, limited percolation	Mainly percolation
Application techniques	Sprinkler, surface or drip	Sprinkler or surface	Usually surface
Need of vegetation	Required	Required	Optional
Climate needs	Winter storage in cold climates	Not critical	Not critical

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779 **Table 2 Previous longevity estimations in different substrates and systems[1]**

Source	Substrate/type systems	Longevity (years)	Land use	Wastewater type	Soil Texture
Drizo et al. (2002)	Electric Arc Furnace (EAF) Steel slag shale	13-37	Batch experiment	Municipal wastewater	-
Dong et al. (2005)	Oyster shell	8-23	Batch experiment	-	-
Xu et al. (2006)	Furnace slag	22	Batch experiment	-	-
Hu et al. (2006)	Soil	29-49-151	Unproductive	~50% mill industry ~25%from other industries and ~25% from domestic waste- water.	Four major types of sandy soil on the site: Rubicon, Roscommon, Augers, and Granby
Heistad et al. (2006)	Filtralite®	5	Sub-surface flow CW	Domestic wastewater	-
Weiss et al. (2008)	Filtralite® Filtra P Soil	5 10 25	-	Domestic wastewater	-
Jenssen et al. (2014)	Infiltration basin	36	-	Domestic wastewater	Glaciofluvial sand and gravel deposit

780 [1] Different volumes, loading and wastewater characteristics assumptions for calculations

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