

1 **Phosphorus fluxes to the environment from mains water leakage: Seasonality and future scenarios**

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3 Ascott, M.J.^a, Goody, D.C.^a, Lapworth, D.J.^a, Davidson, P.^b, Bowes, M.J.^c, Jarvie, H.P.^c and Surridge,
4 B.W.J.^d

5

6 ^a British Geological Survey, Maclean Building, Crowmarsh, Oxfordshire, OX10 8BB, United Kingdom.

7 ^b Environment Agency, Kings Meadow House, Kings Meadow Road, Reading, Berkshire, RG1 8DQ,
8 United Kingdom.

9 ^c Centre for Ecology & Hydrology, Maclean Building, Crowmarsh, Oxfordshire, OX10 8BB, United
10 Kingdom.

11 ^d Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom.

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13 Corresponding Author: Ascott, M.J.*

14 ^a British Geological Survey, Maclean Building, Crowmarsh Gifford, Oxfordshire OX10 8BB, UK.
15 matta@bgs.ac.uk, +44(0)1491 692408

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21 **Abstract**

22 Accurate quantification of sources of phosphorus (P) entering the environment is essential for the
23 management of aquatic ecosystems. P fluxes from mains water leakage (MWL-P) have recently been
24 identified as a potentially significant source of P in urbanised catchments. However, both the
25 temporal dynamics of this flux and the potential future significance relative to P fluxes from
26 wastewater treatment works (WWT-P) remain poorly constrained. Using the River Thames catchment
27 in England as an exemplar, we present the first quantification of both the seasonal dynamics of current
28 MWL-P fluxes and future flux scenarios to 2040, relative to WWT-P loads and to P loads exported from
29 the catchment. The magnitude of the MWL-P flux shows a strong seasonal signal, with pipe burst and
30 leakage events resulting in peak P fluxes in winter (December, January, February) that are >150% of
31 spring (March, April, May)/autumn (September, October, November) fluxes. We estimate that MWL-
32 P is equivalent to up to 20% of WWT-P during peak leakage events. Winter rainfall events control
33 temporal variation in both WWT-P and riverine P fluxes which consequently masks any signal in
34 riverine P fluxes associated with MWL-P. The annual average ratio of MWL-P flux to WWT-P flux is
35 predicted to increase from 15 to 38% between 2015 and 2040, associated with large increases in P
36 removal at wastewater treatment works by 2040 relative to modest reductions in mains water
37 leakage. However, further research is required to understand the fate of MWL-P in the environment.
38 Future P research and management programmes should more fully consider MWL-P and its seasonal
39 dynamics, alongside the likely impacts of this source of P on water quality.

40 **Highlights**

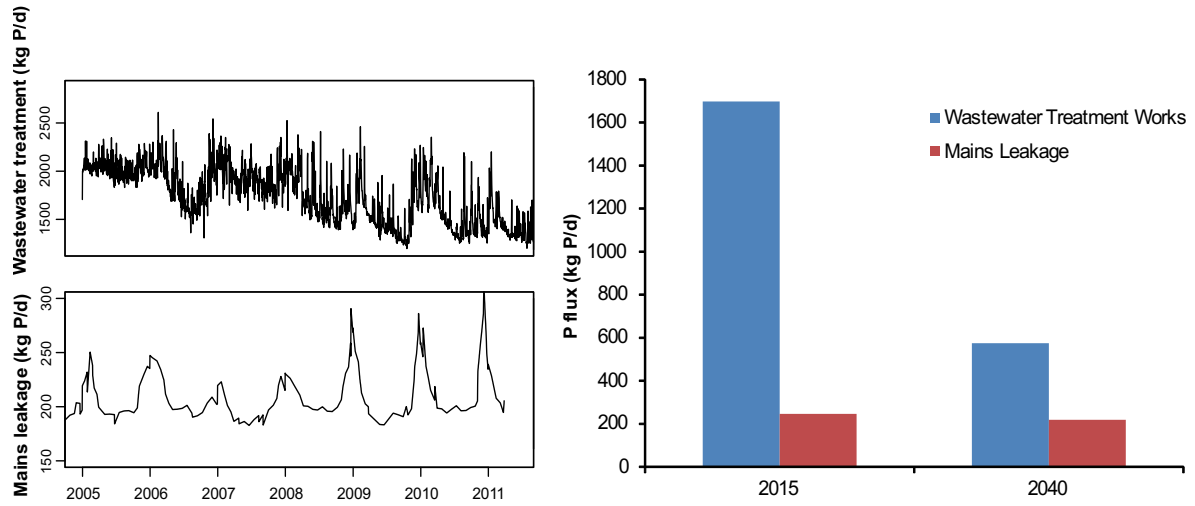
- 41 • Seasonality + scenarios in P fluxes from mains water leakage (MWL-P) quantified
- 42 • MWL-P compared to wastewater P flux (WWT-P) and catchment P export
- 43 • Winter burst MWL-P flux >150% of spring/autumn flux, equal to up to 20% of WWT-P
- 44 • MWL-P/WWT-P ratio predicted to increase to 38% by 2040 due to WWT P removal
- 45 • MWL-P flux and seasonality should be considered in future P research and management

46 **Keywords**

47 Phosphorus, source apportionment, eutrophication, mains water, leakage

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49 **Graphical Abstract**



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59 **1 Introduction**

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61 Eutrophication associated with phosphorus (P) export from agricultural land and with wastewater
62 treatment effluent is a well-known cause of water quality impairment (Elser et al. 2007, Hilton et al.
63 2006, Vollenweider 1968), that can result in harmful algal blooms, fish kills and adverse impacts on
64 animal and human health (Funari and Testai 2008). Effluent from wastewater treatment works has
65 been recognised as a major source of P at the global scale (Morée et al. 2013) which has been shown
66 to impair water quality in rivers in the Europe (Jarvie et al., 2006), Asia (Li et al. 2015), and the USA
67 (Dubrovsky et al. 2010). In the UK, P fluxes from wastewater effluent has been estimated to account
68 for 78% of annual river P loads (White and Hammond 2009). The implementation of the Urban
69 Wastewater Treatment Directive (European Union 1991) resulted in substantial decreases in P loads
70 from UK wastewater treatment works (WWT-P), leading to large decreases in fluvial P fluxes. For
71 example, in 2015, fluvial fluxes of P in England and Wales were estimated to have declined to less than
72 50% of 1974 fluxes (Worrall et al. 2015).

73 Recent studies have shown that in catchments with a large proportion of urban landcover, leakage of
74 P added to mains water (MWL-P) as phosphate is a potentially significant but previously largely
75 overlooked source of P within the environment (Goody et al. 2017). Phosphate is added to mains
76 water to reduce plumbosolvency (Hayes 2010), with the risks associated with lead in drinking water
77 derived from pipework being a significant public health concern (Edwards et al. 2009). Lead
78 consumption in humans has been associated with cognitive development problems in children
79 (Bellinger et al. 1987) as well as increased risk of heart disease and stroke (Pocock et al. 1988).
80 Phosphate is added to mains water to convert lead carbonate to lead phosphate. Lead phosphate is
81 an order of magnitude less soluble than lead carbonate and results in the formation of lead phosphate
82 precipitates on internal pipe surfaces (Hayes 2010). In the UK it is estimated that >95% of water
83 supplies are dosed with phosphate (CIWEM 2011). In the USA, over half of supplies are dosed (Dodrill

84 and Edwards 1995). Dosing achieves final P concentrations in tap water that are estimated to be
85 between 700 – 1900 µg P/L (UK Water Industry Research Ltd 2012), with phosphate dosing having
86 been a highly successful engineering solution to reduce lead concentrations in drinking water. For
87 example, in 2011, 99.0% of random tap water samples in England and Wales met the drinking water
88 limit of 10 µg Pb/L (CIWEM 2011).

89 Leakage of mains water is a globally significant challenge, with the cost of non-revenue water (leakage
90 and unbilled consumption) estimated to be \$14 billion per year (World Bank 2006). National-scale
91 leakage rates in England and Wales are reported to be up to 25% of the water that enters the
92 distribution network and water companies plan future leakage rates over a 25-year planning horizon
93 (2015-2040). Leakage of phosphate-dosed mains water as a potentially important source of P in the
94 environment was first hypothesised by Holman et al. (2008). Goddy et al. (2015) made the first
95 quantification of this flux on an annual basis for England and Wales, which was subsequently refined
96 by Ascott et al. (2016a).

97

98 However, understanding the temporal variability of MWL-P is also important, because biological
99 impacts of P in aquatic ecosystems also show significant temporal variability, with autotrophic
100 biomass growth occurring in many systems primarily during spring (March, April, May) and early
101 summer (June, July, August) (Bowes et al. 2012a). A number of studies (Birek et al. 2014, Cocks and
102 Oakes 2011, UK Water Industry Research Ltd 2007, 2013) have highlighted seasonal dynamics in
103 leakage of water from mains supplies. Higher rates of both bursts and low-level continuous “invisible”
104 leakage occur during winter (December, January, February), due to pipe contraction and expansion.
105 However, work on P to date has only quantified the annual flux of P from mains water leakage and the
106 temporal dynamics of this source are poorly constrained. Whilst substantial reductions in WWT-P
107 loads to rivers have been achieved (Worrall et al. 2015), elevated P concentrations remain a significant
108 concern, with 40% of the water bodies in England not achieving “good status” under the Water

109 Framework Directive (European Union 2000) due to high reactive P concentrations (Environment
110 Agency 2015). Consequently, in England and Wales, substantial further investment in wastewater
111 treatment is planned to 2020 with an estimated total expenditure of £2.1 billion (Global Water Intel
112 2014). Similar programmes exist internationally, for example in the USA (Sewage World 2016) and
113 across Europe (Water Technology 2016). In parallel, water companies continue to reduce leakage
114 based on water saving drivers, which will in turn reduce MWL-P fluxes. Despite these programmes,
115 no estimates of how the relative contributions of MWL-P and WWT-P to the environment may change
116 in the future have been made. If P sources to aquatic ecosystems are to be managed effectively, and
117 the impacts of MWL-P reduced, it is essential that the temporal dynamics of MWL-P and potential
118 future changes in fluxes from this source of P are better constrained. The research reported here was
119 undertaken to examine the following hypotheses:

- 120 1. MWL-P fluxes show seasonal trends, with increased fluxes in winter associated with increased
121 bursts and invisible leakage
- 122 2. Seasonality in MWL-P fluxes can be distinguished from temporal variability in other P sources
123 such as wastewater treatment works effluent
- 124 3. The relative importance of MWL-P fluxes will increase in the future, as WWT-P fluxes decrease
125 due to improvements in tertiary treatment.

126

127 Using the River Thames, a large lowland catchment in the UK, as an exemplar, we tested these
128 hypotheses by deriving historic, daily MWL-P, WWT-P and riverine P fluxes. Subsequently, we
129 validated our approach by comparing our derived annual fluxes of P from MWL and WWT sources to
130 fluxes reported in previous studies. We then undertook time series analysis using multiple linear
131 regression modelling to develop an improved understanding of the temporal dynamics and the
132 processes controlling MWL-P, WWT-P and riverine P fluxes. Further, we derived future scenarios for
133 MWL-P and WWT-P, based on 25 year plans for leakage reduction and WWT-P removal respectively

134 as published by the UK environmental regulator and water companies. Finally, we provide a summary
135 of key research priorities in the context of MWL-P, and consider how to manage this source of P across
136 a range of different hydro-socioeconomic settings (countries which currently undertake phosphate
137 dosing, countries considering this strategy, countries actively avoiding it).

138

139 **2 Materials and Methods**

140 **2.1 Study area**

141

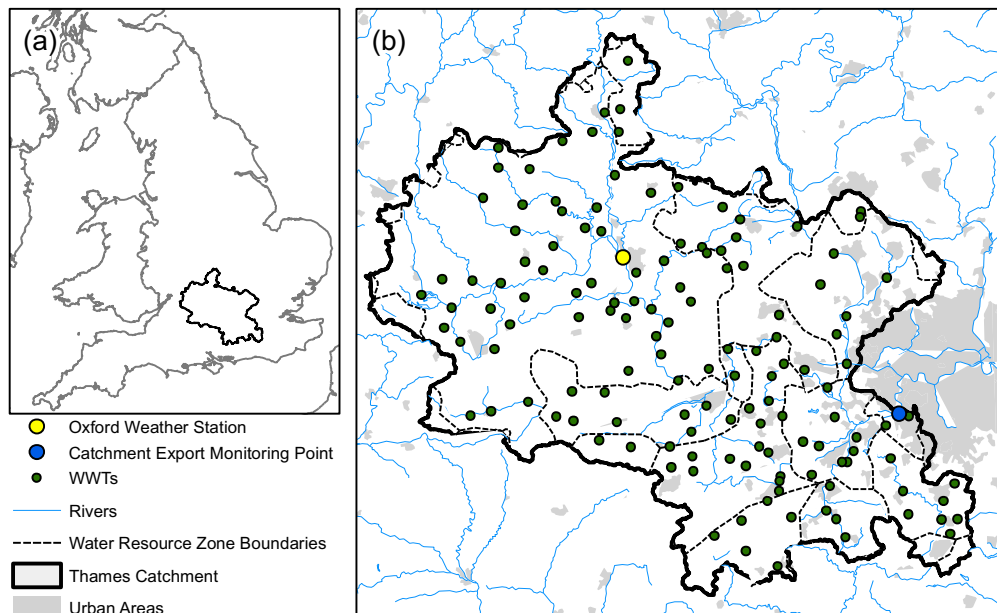
142 We quantified daily MWL-P, WWT-P and riverine P fluxes for the non-tidal Thames catchment (Figure
143 1). The Thames is a relatively large (9948 km²) lowland catchment in southern England. The
144 catchment is predominantly underlain by the Cretaceous Chalk, with Oolitic limestones at the head of
145 the catchment and areas of low permeability clays around Oxford and London. Mean annual rainfall
146 in the catchment at Oxford is 745 mm (Marsh and Hannaford 2008). Although the catchment is
147 relatively rural, with c. 45% of land classified as arable (Fuller et al. 2013), there are also a number of
148 major urban areas (London, Oxford, Reading) resulting in a high catchment population density (c. 960
149 people/km² (Merrett 2007).

150

151 Water supply in the catchment is provided by four different water companies (Thames Water, Affinity
152 Water, Sutton and East Surrey Water and South East Water) which operate across a total of 10 largely
153 self-contained water resource zones (WRZs, Environment Agency (2012)). Each individual water
154 company reports annual leakage rates. Leakage rates are reported to be high, reaching up to 26% of
155 water input to the distribution network (Committee on Climate Change 2015). The catchment has
156 been subject to previous research to estimate the annual contribution of MWL-P and WWT-P to the

157 River Thames (Goody et al. 2017). Building on this initial research, Ascott et al. (2016a) estimated
158 that c. 30% of the MWL-P flux in England and Wales was derived from the river basin district in which
159 the non-tidal Thames catchment is located. Wastewater treatment occurs throughout the catchment
160 and 137 water company wastewater treatment works serving > 2000 population equivalents (p.e) are
161 present. Since the late 1990s, P fluxes from WWTs have been reduced in the catchment through the
162 introduction of tertiary treatment, primarily driven by the EU Urban Waste Water Treatment Directive
163 (Kinniburgh and Barnett 2010, Powers et al. 2016). Consequently, P concentrations in the River
164 Thames fell by c. 90% between 1992 and 2005 (Kinniburgh and Barnett 2010). Despite this, there is
165 scant evidence that eutrophication risk in the Thames has reduced (Bowes et al. 2014), with a number
166 of rivers in the catchment exhibiting excessive phytoplankton biomass and P concentrations above
167 those at which the growth of autotrophs can be expected (> c. 80 µg SRP/l, Bowes et al. (2012b)).

168



169

170 **Figure 1** Location of the non-tidal Thames catchment within England (a) and the water resource zones, wastewater
171 treatment works serving > 2000 p.e., catchment export monitoring point and the Oxford weather station within the
172 catchment (b). Contains Ordnance Survey data © Crown copyright and database right (2017).

173

174

175 **2.2 P flux time series derivation and analysis**

176 **2.2.1 Data sources**

177

178 Table 1 summarises the data sources used to derive daily time series of MWL-P, WWT-P and riverine
 179 P fluxes in this study for the period 2001 - 2011. MWL-P was estimated based on published leakage
 180 rates and estimates of P concentrations in drinking water. Water quality sampling of WWT discharges
 181 and the River Thames occurs at a much lower frequency (c. every 10 days to monthly) in comparison
 182 to the daily flow data that are available. Consequently, to derive daily WWT-P and riverine P fluxes,
 183 missing concentration data were estimated using concentration-flow relationships, as detailed in
 184 section 2.2.3.

185

186 **Table 1 Data sources used to derive daily time series of MWL-P, WWT-P and riverine P fluxes for 2001 - 2011.**

Flux	Data	Time period	Frequency	Source
MWL-P	WRZ level leakage rates for WRZs in non-tidal Thames catchment	2011 - 2013	Annual	WRMP tables
	Leakage rates for Thames Water	2001 - 2011	Daily	Cocks and Oakes (2011)
	PO ₄ -P dosing concentration	2001 - 2011	Annual	Comber et al. (2011)
	PO ₄ -P dosing extent	2001 - 2011	Annual	Hayes et al. (2008), CIWEM (2011)
Riverine P	PO ₄ -P concentration for Thames at Teddington	2000 - 2015	Monthly	Environment Agency
	Flows for the Thames at Teddington	2000 - 2015	Daily	
WWT-P	PO ₄ -P WWT discharge concentrations	2005 - 2011	Variable (mean frequency = 10 days, median frequency = 27 days)	Environment Agency
	WWT discharge flows	2005 - 2011	Daily	

187

188

189

190 **2.2.2 MWL-P**

191

192 Annual water resource zone level leakage rates (in ML/day) for 2011-2013 were extracted from water
193 resources planning tables available on water company websites (Affinity Water 2014, Southeast Water
194 2014, Sutton and East Surrey Water 2014, Thames Water 2014). Daily leakage data for 2001 to 2011
195 (as reported by Cocks and Oakes (2011)) for the whole Thames Water supply area were used as the
196 basis for estimating the MWL-P flux. This is the only publicly-available sub-annual leakage data for
197 both the catchment and the UK. The Thames Water supply area covers 76% of the non-tidal Thames
198 catchment that is the focus for the research reported here, but also includes London in the
199 downstream tidal Thames catchment. To account for this discrepancy, we calculated the ratio of the
200 annual leakage rates in the water resources planning tables for the whole Thames Water supply area
201 versus the 10 WRZs in the non-tidal Thames catchment. We then used this ratio to scale the daily
202 Thames Water leakage time series (Cocks and Oakes 2011) to provide a daily leakage time series for
203 the non-tidal Thames catchment to Teddington for the same period. Over the period 2001 to 2011,
204 the extent of P dosing of mains water and the concentrations used have increased. Based on published
205 reports of dosing concentrations (Comber et al. 2011) and the spatial extent of dosing (CIWEM 2011,
206 Hayes et al. 2008, UK Water Industry Research Ltd 2012), we used the conservative approach adopted
207 by Goody et al. (2017) to estimate the historic changes in P dosing concentrations and extents within
208 the non-tidal Thames catchment.

209

210 **2.2.3 Riverine P and WWT-P flux derivation, flux comparison and time series analysis**

211

212 *Riverine P flux derivation*

213

214 The daily net export of P (2001-2016) from the catchment was derived using observed
215 orthophosphate-P ($\text{PO}_4\text{-P}$) concentration and daily flow data for the River Thames at Teddington
216 (Figure 1). In order to infill missing concentration data to derive a daily P flux time series, we first
217 undertook single change point detection using an asymptotic penalty value of 0.05 (Chen and Gupta
218 2011, Killick and Eckley 2014) to determine whether there was a statistically significant change in the
219 mean and variance of the concentration time series associated with historic reductions in P loads from
220 WWTs (Kinniburgh and Barnett 2010). Following the approach of Bowes et al. (2014), we then used
221 non-linear least-squares regression to derive separate concentration-flow relationships before and
222 after any change points that were identified:

$$223 \qquad C = AQ^B \qquad (1)$$

224 Where C is concentration (mg P/L), Q is flow (m^3/s) and A and B are empirically derived constants. The
225 derived relationships were used to estimate P concentrations where data were missing before and
226 after the change point. The complete concentration and flow time series was then used to derive a
227 daily P flux time series.

228

229 *Wastewater Treatment Works P Flux Estimation*

230

231 Wastewater treatment works in the Thames catchment fall into two categories: (1) monitored sites
232 where flows are $> 50\text{m}^3/\text{d}$; and (2) unmonitored, small sites with flows $< 50\text{m}^3/\text{d}$. Outputs from the
233 Thames Source Apportionment Geographical Information Systems (SAGIS, Comber et al. (2013)) tool
234 using population equivalent data and concentration estimates have shown that small unmonitored
235 sites contribute $< 1\%$ of the total WWT-P load to the River Thames. Consequently, these sites have not
236 been considered further in the research reported here.

237

238 For the monitored WWT sites, daily flow monitoring data for 2005 – 2011 were used in conjunction
239 with PO₄-P concentration data to derive daily P fluxes for each site. A similar approach to that
240 described above for the riverine P flux estimates was used for infilling of missing concentration data
241 using concentration-flow relationships of the form of equation 1. The dates for implementation of P
242 removal schemes at WWTs in the Thames catchment were provided by the environmental regulator
243 for England. Where P removal schemes were implemented at a WWT during the study period,
244 separate concentration-flow relationships were estimated for before and after implementation.
245 Where no changes in treatment processes for P removal at WWTs occurred during the study period,
246 a single concentration-flow relationship for the WWT was used. The derived daily P fluxes for each
247 individual WWT were then summed to derive a total, daily catchment flux of WWT-P.

248

249 *Flux comparison and time series analysis*

250

251 To validate our daily estimates of MWL-P and WWT-P, we derived annual mean fluxes for each of
252 these P sources and then compared these to previously published estimates of the same fluxes for the
253 Thames catchment (Comber et al. 2013, Goody et al. 2017, Haygarth et al. 2014). We also calculated
254 a daily time series of the ratio of MWL-P to WWT-P. The derived MWL-P, WWT-P and riverine P time
255 series were initially compared by visual inspection to assess the timing and magnitude of peaks in
256 fluxes. We then assessed whether there were significant long term trends and changes in the variance
257 in MWL-P and MWL-P/WWT-P ratio using a Mann Kendall test (Pohlert 2018) and a change point
258 analysis using an asymptotic penalty value = 0.01 (Killick and Eckley 2014).

259

260 In order to assess the control of cold weather events on temporal variability in MWL-P, we undertook
261 correlation analyses using historic daily minimum temperature data for the Oxford station in the

262 Thames catchment (Figure 1). We also quantified correlations between daily rainfall and daily WWT-
263 P and daily riverine P loads to assess the role of short term rainfall events in controlling P fluxes.
264 Finally, to determine how much of the variation in daily riverine P load could be explained by daily
265 MWL-P and daily WWT-P, we developed simple multiple linear regression models using (1) WWT-P
266 only and (2) WWT-P + MWL-P as explanatory variables. We compared the coefficient of determination
267 between the two models and used a partial F-test (R Development Core Team 2016) to determine the
268 impact of adding the MWL-P variable on model predictive power. There is substantial uncertainty in
269 the fate of MWL-P in the environment associated with the relative importance of different P retention
270 (clay sorption, soil and sediment accumulation) and transport processes (groundwater, surface water
271 and the sewer network) (Goody et al. 2017). In this context, our research does not aim to make a
272 direct causal link between time series variability in WWT-P, MWL-P and riverine P fluxes. The purpose
273 the comparison of fluxes described here is threefold: (1) to provide indications of the relative
274 contributions of these sources to the environment; (2) to improve understanding of the processes
275 controlling temporal variability in these sources; and (3) to improve understanding of the relationships
276 between the sources.

277

278

279 **2.3 Derivation of future scenarios of MWL-P and WWT-P**

280

281 Scenarios for future reductions in both MWL-P and WWT-P were constructed. Planned leakage
282 reductions for each of the 10 WRZs in the Thames catchment for 2015-2040 were extracted from
283 water company water resource planning tables (Affinity Water 2014, Southeast Water 2014, Sutton
284 and East Surrey Water 2014, Thames Water 2014). Estimates of future reductions in WWT-P were
285 provided by the environmental regulator as new discharge permit limits for P (in mg P/L). In England
286 and Wales investments in wastewater treatment are planned on a five-year Asset Management

287 Programme (AMP) cycle. Investments over 2015 – 2020 (Asset Management Programme 6 (AMP6))
288 are fully planned and costed. Further into the future, a provisional “long list” of improvements in
289 WWTs that are potentially required in order to meet good ecological status in the catchment has been
290 derived based on source apportionment modelling (Comber et al. 2013). This list has been subject to
291 a cost benefit analysis which has divided sites into those at which enhanced P removal is cost beneficial
292 and those at which this is not. In total, four WWT improvement scenarios were developed, as detailed
293 below:

- 294 1. Baseline (2015)
- 295 2. AMP6 (2020) – planned and costed WWT improvements and leakage reduction programmes
296 for 2015 – 2020
- 297 3. Cost beneficial planning period (2040a) – planned leakage reductions and cost beneficial WWT
298 improvements for 2015 – 2040
- 299 4. Full planning period (2040b) – planned leakage reductions and all potential WWT
300 improvements for 2015 – 2040

301 Scenarios were implemented by applying the new discharge permit limits for WWTs to the baseline
302 observed concentration-flow data in 2015. Given the uncertainty in future WWT and leakage
303 reduction beyond 2020, an annual average approach was used to derive annual fluxes from MWL-P
304 and WWT-P in 2015, 2020 and the two 2040 scenarios.

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309 **3 Results**

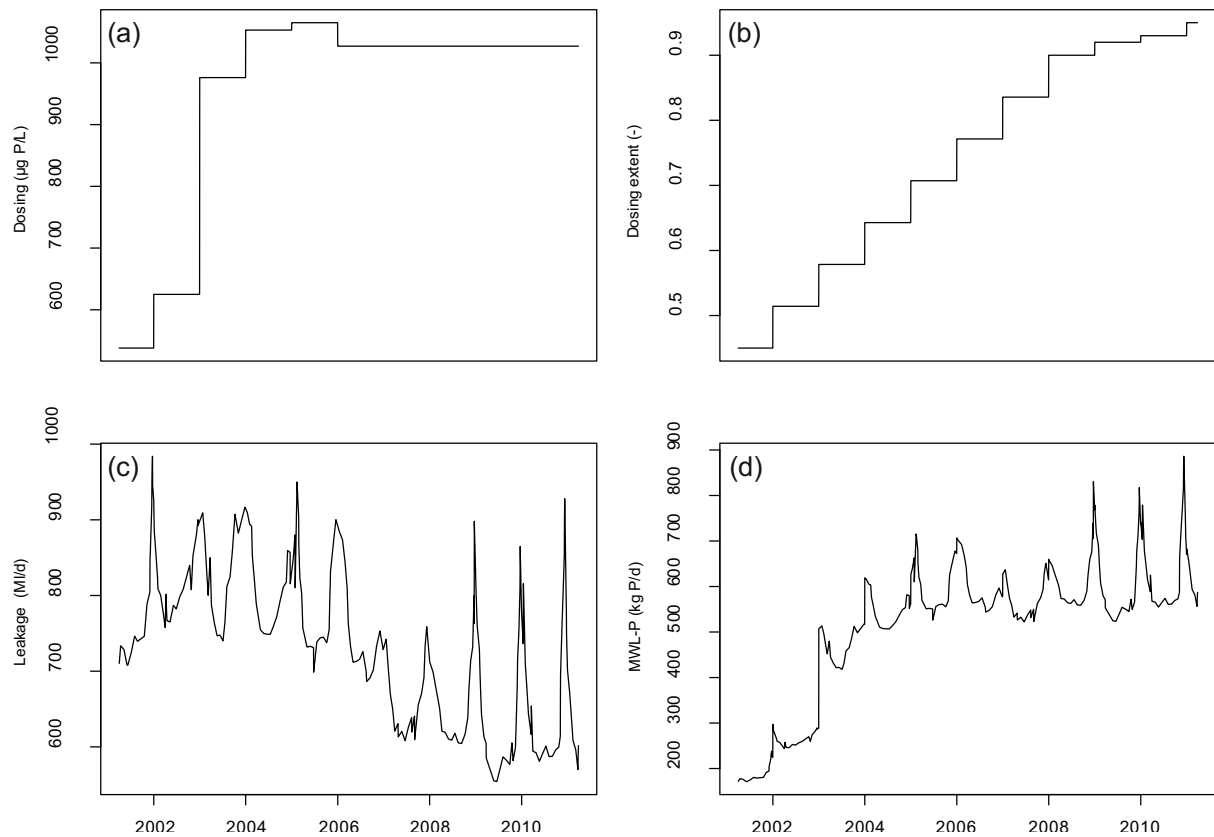
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311 **3.1 MWL-P flux estimation**

312

313 Figure 2 shows the evolution of the MWL-P flux over the period 2001 to 2011 for the Thames
314 catchment. Figure 2 (c) shows the total daily leakage (Cocks and Oakes 2011) for 2001 – 2011 for
315 Thames Water. Over the period 2001 – 2011, the volume of leakage has declined due to water
316 companies actively locating and fixing leaks in the distribution network. After accounting for this
317 reduction, the leakage time series shows a relatively strong correlation at lag = 0 with daily minimum
318 temperature (Pearson correlation, $r = 0.63$, $p = 1.392 \times 10^{-14}$). The time series shows a clear seasonal
319 signal associated with increased bursts and invisible leakage in winter. The winters of 2009, 2010 and
320 2011 were particularly cold, with mean December temperatures 1.1, 2.0 and 4.8 degrees below long
321 term average (1971-2000) respectively (Met Office 2016). This resulted in large increases in leakage
322 of up to 50% compared to the preceding autumn months (September, October, November). In
323 summer (June, July, August), small increases of up to 6% compared to spring (March, April, May) values
324 are also observed, which has been attributed to soil expansion and contraction resulting in pipe failure
325 (Cocks and Oakes 2011). Figure 2 (d) shows the estimated daily flux of MWL-P for 2001-2011 for the
326 Thames catchment. Whilst the volume of leakage has reduced substantially, increases in the spatial
327 extent of dosing and in phosphate dosing concentrations counteract these reductions, resulting in
328 significant (Mann Kendall trend test, $p < 2.2 \times 10^{-16}$) net increases in MWL-P fluxes of c. 300% over the
329 period 2001 to 2011. Seasonality increases through time with a significant change in variance
330 identified at 08/09/2008 (single change point detection using an asymptotic penalty value =0.01) and
331 the ratio of daily annual maximum/minimum MWL-P rates increasing from c. 130% in 2002 – 2008 to
332 150% in 2008 – 2011.

333



334

335 **Figure 2: Estimates of P dosing concentrations (a) and extents (b) for 2001 – 2011, daily leakage rates reported by Cocks**
 336 **and Oakes (2011) (c), and the derived MWL-P flux (d)**

337

338

339

340 **3.2 WWT-P flux estimation**

341

342 Figure 3 reports data related to the WWT-P flux for the Thames catchment. Substantial decreases in

343 P concentration in WWT effluents occurred between 2005 and 2007, associated with the introduction

344 of P removal technology. From March 2008 to 2012, no significant further P removal was implemented

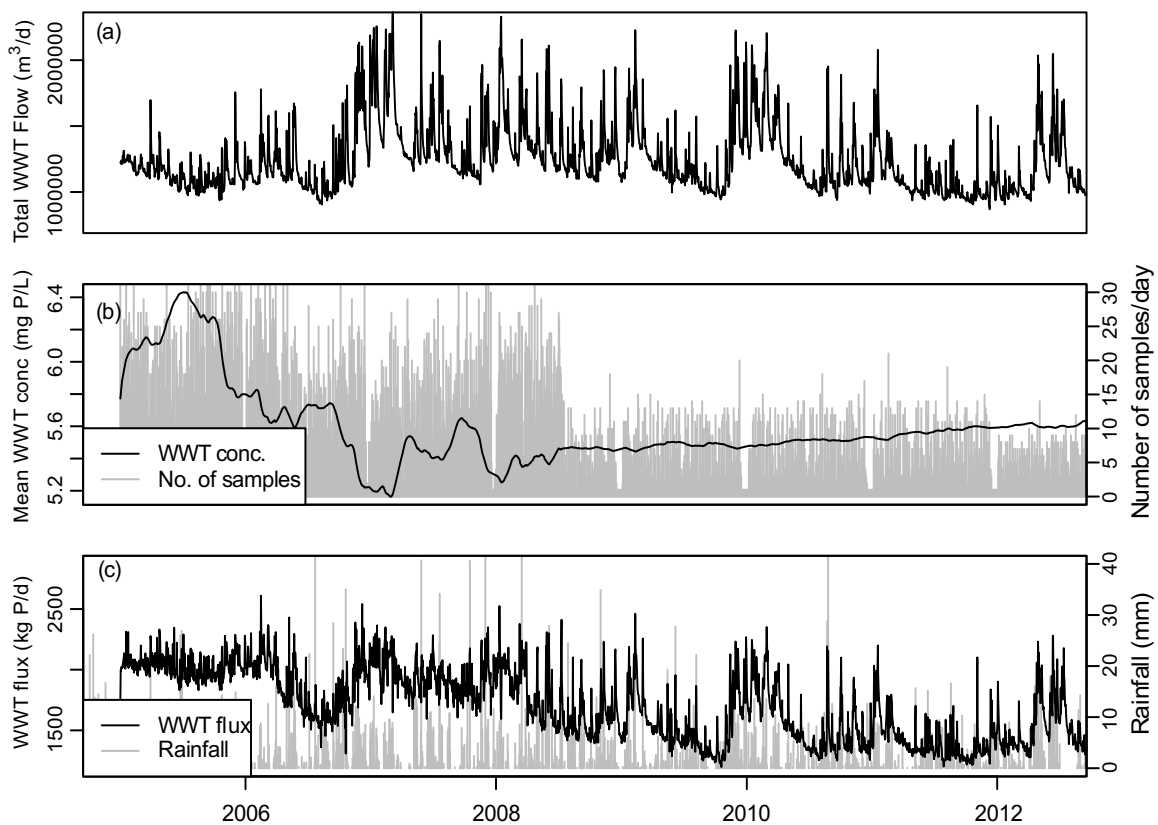
345 in the catchment. Consequently, effluent P concentrations have been relatively stable and the

346 frequency of P monitoring reduced. The slight increase (c. 5.4 to 5.6 mg P/L) in effluent P

347 concentrations over this period may reflect an overall decrease in P removal efficiency in existing

348 WWTs due to changes in plant operation and influent P loads (Tchobanoglous and Burton 2013). There

349 is considerable variation in the WWT flows (Figure 3 (a)) particularly during high flow periods in winter
 350 associated with rainfall events when flows can be up to twice as large as dry weather flows (DWF).
 351 The derived WWT-P flux (Figure 3 (c)) reflects both long term changes in P concentrations following
 352 investment in P removal technologies and short term variability in WWT flows associated with rainfall
 353 events.
 354



355
 356 **Figure 3 Total WWT flows (a), mean daily P concentrations and number of samples (b) and the derived WWT-P flux and**
 357 **rainfall time series for Oxford (c)**

358
 359 **3.3 Flux comparison**

360
 361 Table 2 reports the annual mean MWL-P and WWT-P fluxes derived in the research reported here and
 362 in previous studies of the Thames catchment. The annual estimates of MWL-P in this study are within
 363 the range of values reported by Goody et al. (2017) who considered a range of different plausible

364 historic mains water dosing extents. WWT-P fluxes broadly corroborate previous findings reported by
 365 Haygarth et al. (2014) and Comber et al. (2013). Fluxes estimated in the current study for 2006-2008
 366 are somewhat higher (c. 29%) than estimates made using observed soluble reactive P concentrations
 367 in WWT discharges in conjunction with source apportionment tools (Comber et al. 2013). Fluxes from
 368 the source apportionment modelling were adjusted by the UK environmental regulator (Environment
 369 Agency 2016) to take into account new and enhanced P removal which is the likely cause of this
 370 discrepancy. Our estimates are also larger (c. 50%) than those calculated by Haygarth et al. (2014),
 371 who used a simple approach based on population equivalents and assumed total P concentrations to
 372 estimate WWT-P flux for the catchment.

373 **Table 2 Annual mean MWL-P and WWT-P fluxes derived in this study and previous studies**

374

Period	MWL-P (kt P/yr)		WWT-P (kt P/yr)		
	Gooddy et al. 2017	This study	Haygarth et al 2014	Comber et al 2013	This study
2001	0.019 - 0.029	0.025	1.18	-	-
2006-2008	-		-	0.52	0.67
2011	0.068 - 0.089	0.082	0.38	-	0.58

375

376 **3.4 Flux comparison between MWL-P and WWT-P with Riverine P Export**

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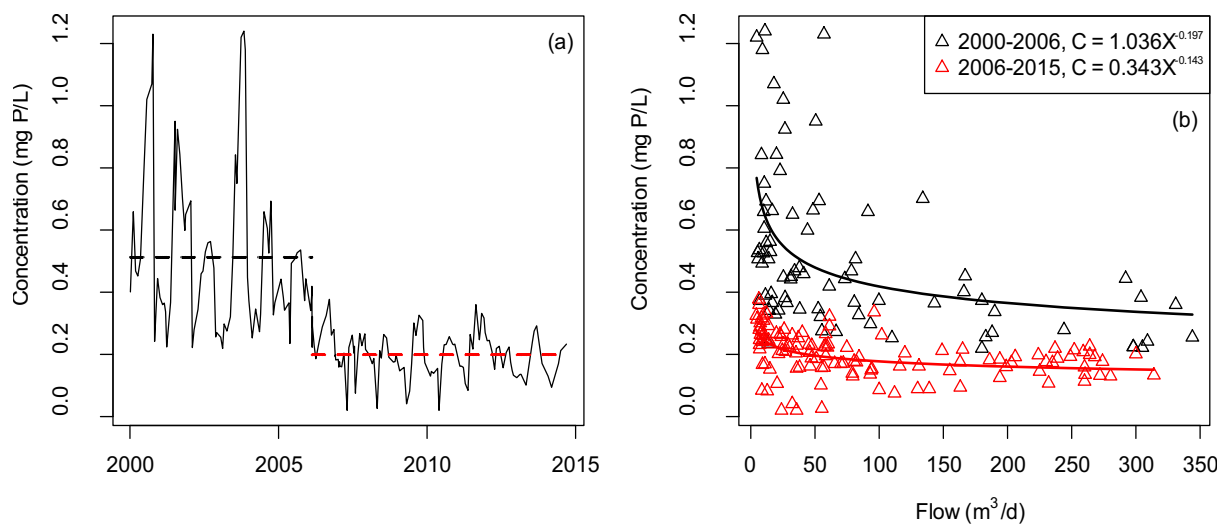
378 **3.4.1 Riverine P flux estimation**

379

380 Figure 4 shows the results of the change point detection analysis and the derived concentration-flow
 381 relationships before and after the change point for the River Thames at Teddington. A substantial
 382 decrease in the mean and variance of riverine phosphate concentrations was observed in 2006,
 383 associated with reductions in P loads from WWTs. For the period 2000 – 2006, the concentration-flow
 384 relationship shows decreases in concentrations with increasing flow, associated with dilution of WWT
 385 inputs. This corroborates concentration flow relationships derived by Bowes et al. (2014) in the

386 Thames basin. From 2006 to 2015, the variability in concentration with flow is substantially reduced.
 387 Between 0 and 100 m³/d decreases in concentration with increasing flow are observed, associated
 388 with dilution of point source P inputs. However, from 100 to 500 m³/d, there is limited change in
 389 concentration with increasing flow which reflects a balance of both point source dilution and
 390 mobilisation of diffuse P sources across this range of discharges.

391



392

393 **Figure 4 Observed phosphate concentrations (mg P/L) in the River Thames at Teddington and mean concentrations**
 394 **(dashed line) before and after the change point (a) and concentration-flow relationships for each of these periods (b).**

395

396

397 **3.4.2 Comparison of MWL-P, WWT-P and riverine P fluxes**

398

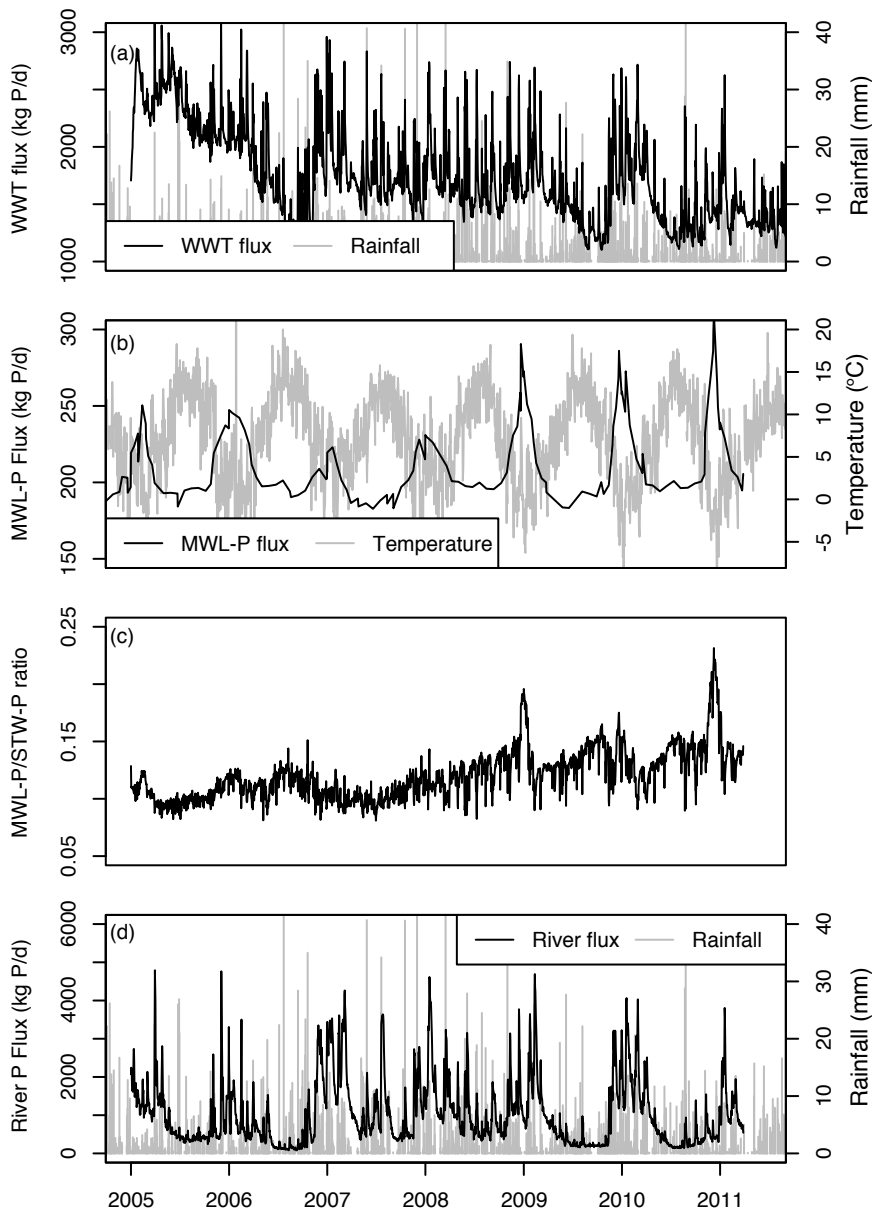
399 Figure 5 shows the ratio of MWL-P and WWT-P fluxes (“MWL-P/WWT-P”) and their relationship with
 400 P flux in the River Thames at Teddington. From 2005 to 2011 there is a significant trend in MWL-
 401 P/WWT-P (Mann-Kendall trend test, $p < 2.2 \times 10^{-16}$). The ratio of MWL-P to WWT-P increases from
 402 from c. 7-10 in 2005 to 15% in 2011. This is due to two factors: (1) an increase in the extent of mains
 403 water P dosing through the period, as shown in Figure 2; and (2) a decrease in WWT-P loads due to
 404 investment in P removal technology.

405 Seasonality in the ratio of MWL-P to WWT-P increases through the period, due to the increased extent
406 of dosing. There is a significant increase in the variance of MWL-P/WWT-P from 22/11/2008 onwards
407 (single change point detection using an asymptotic penalty value = 0.01 (Chen and Gupta 2011, Killick
408 and Eckley 2014)). Peaks in MWL-P/WWT-P of up to 20% occur in the winters of 2009 and 2011 in
409 comparison to values around 10% in 2005-2008. This seasonality also gives some initial insight into
410 the processes controlling MWL-P fluxes. In the winter of 2009/10 a rapid increase in MWL-P occurs
411 followed by a rapid increase in WWT-P. This causes MWL-P/WWT-P to rise and fall again rapidly. It is
412 likely these trends are the result of a particularly cold weather period resulting in an increase in leaks
413 and bursts. This is likely to be followed by a period of active leakage control to reduce leakage rates
414 back to levels prior to the cold weather period, as has been reported by UK Water Industry Research
415 Ltd (2007). At the same time a period of wet weather occurs which results in increased inflows to
416 WWTs. This results in increases in WWT outflows (Figure 3 (a)) but flow increases are significantly less
417 than typical design flows for full treatment of influent during storms (up to 3 times DWF, with 4 – 6
418 times DWF retained in storm tanks (Saul et al. 2007)). Any influent in excess of storm tank design
419 criteria as well as combined sewer overflows are likely to be discharged directly to the nearest
420 watercourse. These will all combine to reduce the effectiveness of the WWTs to remove P.

421

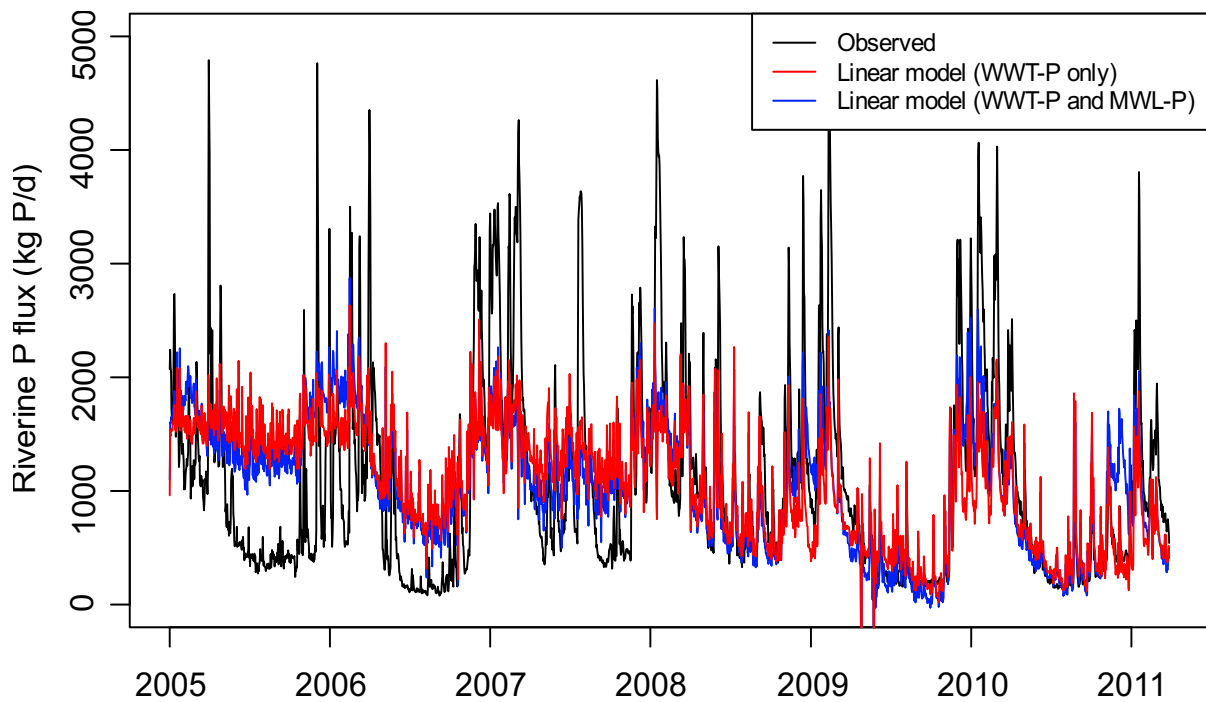
422 Figure 5 (a) and (d) show the daily WWT-P flux and catchment P export. There are weak but significant
423 correlations between WWT-P and daily rainfall (Pearson correlation, $r = 0.23$, $p < 2.2 \times 10^{-16}$) and
424 catchment export and daily rainfall (Pearson correlation, $r = 0.18$, $p < 2.2 \times 10^{-16}$). Figure 6 shows
425 observed and modelled riverine P fluxes using MLR models considering: (1) WWT-P only; and (2) WWT-
426 P and MWL-P together. The multiple linear regression model used to determine whether the riverine
427 P load can be explained by MWL-P and WWT-P resulted in a relatively modest coefficient of
428 determination (adjusted $R^2 = 0.41$). The MLR model suggested that WWT-P and MWL-P are both
429 significant explanatory variables ($P < 2 \times 10^{-16}$). Removing MWL-P from the regression significantly

430 reduced the predictive power of the model ($F = 242$, $P < 2 \times 10^{-16}$, for partial F-test between MLR
431 models considering WWT-P only and WWT-P + MWL-P) resulting in a lower coefficient of
432 determination (adjusted $R^2 = 0.33$).



433

434 **Figure 5** Daily fluxes of WWT-P and rainfall (a), MWL-P and temperature (b) for the Thames catchment, the ratio of MWL-
435 P to WWT-P (c) and the riverine flux of P out of the catchment and rainfall (d)



436

437 **Figure 6: Observed (black) and modelled riverine P fluxes for the export of the Thames catchment using the MLR model**
 438 **considering WWT-P only (blue) and WWT-P and MWL-P (red)**

439

440

441 **3.5 Future loads of MWL-P and WWT-P**

442

443 Table 3 reports the derived future MWL-P and WWT-P loads for annual average conditions. WWT-P
 444 fluxes are predicted to decrease to 2020 associated with the AMP6 programme, with further
 445 decreases predicted to 2040. When considering all WWT improvements to 2040, a total decrease in
 446 WWT-P flux of 0.41 kt P/yr is estimated. Small reductions in MWL-P of 0.01 kt P/year are predicted to
 447 occur to 2040. In total, the reduction in MWL-P loading is c. 2% of the reduction in WWT-P.
 448 Consequently, the ratio of MWL-P to WWT-P is predicted to increase from c. 15% to 38% under
 449 average conditions.

450

451

452

453 **Table 3 Future MWL-P and WWT-P loadings**

454

Scenario Information				Fluxes		MWL-P/WWT-P %
Scenario Name	Time Slice	WWT Improvements	Leakage Improvements	WWT-P Flux (kt P/yr)	MWL-P Flux (kt P/yr)	Annual Mean
Baseline	2015	-	-	0.62	0.09	14.52
AMP6 Programme	2020	AMP6 Programme	AMP6 Leakage Programme	0.61	0.08	13.11
2040 Plans (Cost Beneficial)	2040	CBA "Long List"	2040 Plan Leakage Programme	0.57	0.08	14.04
2040 Plans (All sites)		Full "Long List"		0.21	0.08	38.10
Total P flux improvement (kt P/yr)				0.41	0.01	

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463 **4 Discussion**

464 **4.1 Seasonal dynamics and relative importance of MWL-P and WWT-P to** 465 **river P loads**

466

467 Estimates of MWL-P and WWT-P fluxes for the Thames catchment made in the research reported here
468 illustrate that MWL-P is a potentially significant source of P in the environment which has so far been
469 largely overlooked in P source apportionment and management strategies. In particular, the distinct
470 seasonal signal (Figure 2) in MWL-P results in higher ratios of MWL-P to WWT-P through the winter
471 periods. The seasonality in MWL-P that was hypothesized in section 1 has not been reported to date
472 and represents a development in our understanding of this P source.

473

474 The processes controlling seasonality in MWL-P are complex. Whilst correlations between minimum
475 air temperatures and MWL-P are reported in this paper, it should be noted that other non-
476 meteorological processes also have a potentially significant impact on MWL-P variability. Cold weather
477 is the predominant control on the timing and magnitude of the initial outbreak of water mains bursts
478 during a winter leakage event. As temperatures increase, there is likely to be some natural reduction
479 in invisible leakage associated with pipe expansion. However, rather than any meteorological factor,
480 the majority of the reduction in leakage after an event is the result of active repairs to burst water
481 mains (UK Water Industry Research Ltd 2007). This combination of meteorological and engineering
482 processes is likely to be broadly applicable in developed countries with significant temperature
483 fluctuations and consequently a similar seasonality in MWL-P would be expected. Phosphate dosing
484 is starting to be developed in other temperate countries (e.g. Ireland (Irish Water 2015, Mockler et al.
485 2017)). In these countries, as dosing extents and concentrations increase, an increase in the both the
486 absolute magnitude and seasonality of MWL-P fluxes, as reported in Figure 2, would be anticipated.

487

488 Rainfall events have an impact on the observed correlations between WWT-P, MWL-P and riverine P
489 fluxes. Winter rainfall events cause short term increases in both WWT-P and riverine P fluxes but
490 through different processes. High rainfall results in increased inflows to wastewater treatment works.
491 There may be some dilution of influent P during these events due to increased contributions from
492 runoff and groundwater infiltration (De Bénédittis and Bertrand-Krajewski 2005), but these are
493 unlikely to be captured in the flux estimates due to the paucity of concentration measurements.
494 Influent flows below the peak storm flow treatment design criteria (typically 3 x DWF (Saul et al. 2007))
495 will be treated and discharged to watercourses. Excess flows will be temporarily diverted to storm
496 tanks and flows > 6 x DWF will be directly discharged to rivers. Winter rainfall events can cause
497 increases in riverine P fluxes through a number of processes. Increased agricultural runoff, in-stream
498 mobilisation of P associated with sediments, combined sewer overflows (CSOs) and wastewater
499 treatment works discharges can all contribute to increasing P loads during winter storm events (Bowes
500 et al. 2008, Jarvie et al. 2006). These processes controlling short term temporal changes in both
501 riverine P fluxes and WWT-P fluxes mask any correlation between MWL-P and riverine P.

502

503 In the context of MWL-P, there is some uncertainty in the extent of P dosing and the concentrations
504 used. Just as water companies publically release water resource planning data, making historic P
505 concentrations and dosing extents available would be beneficial to refining MWL-P estimates. Given
506 the potential significance of this flux, assessment of MWL-P using observed leakage and P
507 concentration data at the local scale using district metered area (DMA) data could provide important
508 new insights. Further, the uncertainty surrounding the ultimate fate of MWL-P following leakage is
509 significant. As discussed previously, comparisons with WWT-P and riverine P fluxes such as those
510 presented in Figure 5 are beneficial as they provide indications of the relative contribution of different
511 sources to the environment. It is likely that MWL-P makes some direct contribution to riverine P fluxes,

512 but the relative importance of groundwater, surface water and the sewer network as pathways for
513 MWLP-P are largely unknown at this time. Moreover, the spring and summer growing period is often
514 perceived to be the most biologically-critical period in many receiving waters (Jarvie et al. 2006),
515 meaning that winter peaks in MWL-P which do directly contribute to riverine P loads may have a
516 limited immediate impact on riverine ecosystems. However, there are likely to be time lags between
517 the flux of P from a water mains leak and this flux reaching a river, with a range of transit times
518 depending on catchment size, transport pathway and change in P concentration along the pathway.
519 Consequently, it is plausible that winter peaks in leakage of P from water mains only reach a river
520 network in more biologically-critical periods of the year. Long transit times are likely in catchments
521 with a significantly thick unsaturated zone and long groundwater flow paths. In these catchments it
522 is plausible that MWL-P contributes to legacy P stores in groundwater (Holman et al. 2008) as has
523 been observed for nitrate (Ascott et al. 2017, Ascott et al. 2016b). River sediments can also
524 accumulate P (Sharpley et al. 2013), with time lags of < 1 year associated with in-stream P
525 remobilisation during high river flow events. These in-stream accumulation-remobilisation processes
526 will result in further lags between MWL-P that reaches a river and P riverine exports. Whilst not
527 considered in this study, agriculture remains a significant P source to the environment (White and
528 Hammond 2009) and should clearly be accounted for in future P management strategies. Much of the
529 likely temporal dynamics of the fate of MWL-P (higher winter fluxes with potential catchment legacy
530 stores and lags) are analogous to the fate of nonpoint P sources such as agriculture (Sharpley 2016).
531 The anticipated impact of measures to reduce P fluxes from both mains leakage and agriculture should
532 be tempered with knowledge that catchment lags and release of existing nonpoint legacy P sources
533 may significantly delay any water quality improvements.

534

535

536

537 **4.2 MWL-P and WWT-P Scenarios: Implications for drinking water and** 538 **wastewater management**

539

540 Table 3 illustrates that future WWT-P reductions are likely to be substantially larger than MWL-P
541 reductions, and consequently this will result in a greater relative contribution of MWL-P to P loads
542 delivered into the environment in the future. Whilst policy responses to minimise MWL-P were
543 previously discussed by Goody et al. (2017), the implications of future increases in the relative
544 contribution of MWL-P have not been considered to date. Such increases are likely to occur in
545 countries where P dosing is currently being considered in future lead reduction strategies (e.g. Ireland
546 (Irish Water 2015) and South Korea (Lee, pers. comm.)). In these countries, the potential impacts of
547 MWL-P should be considered in environmental impact assessments. Moreover, in less developed
548 countries, current wastewater P removal and mains water P dosing may be less extensive. As both
549 environmental and public health standards improve, it is plausible that in these countries mains water
550 P dosing and WWT P removal may increase substantially, resulting in increases in the relative
551 importance of MWL-P.

552

553 A number of European countries have adopted policies that actively avoid phosphate dosing on
554 environmental grounds (CIWEM 2011). Countries such as the Netherlands undertake pH correction
555 and centralized water softening to ensure lead concentrations are below the required standard (Hayes
556 2012). P has been shown to be a limiting nutrient to biofilm growth in a number of different drinking
557 water systems (Liu et al. (2016) and references therein) and concern has been raised historically that
558 the presence of P in water mains (through dosing or otherwise) would result in increased bacterial
559 counts in water mains (Miettinen et al. 1996). The current and future significance of MWL-P in

560 comparison to other P sources identified in this study adds to the body of evidence to support policies
561 that avoid phosphate dosing, where the extent of lead piping in the distribution network is small
562 enough to safeguard human health protected without dosing.

563

564 Across these different hydro-socioeconomic settings, future management of MWL-P and WWT-P is
565 likely to be a significant challenge. Both water utilities and environmental regulators have often
566 historically been divided between clean water and wastewater (Ofwat 2017). Within the clean water
567 sector, the industry has often been further divided between drinking water quality (responsible for P
568 dosing) and water resources management (responsible for leakage) (Deloitte 2014). Moreover,
569 regulatory drivers for changes in water management have typically addressed single issues (e.g.
570 drinking water quality via the EU Drinking Water Directive (European Commission 1998),
571 environmental quality via the EU Water Framework Directive (European Union 2000)), whereas
572 addressing MWL-P requires policy interventions across multiple fields. Where P dosing and WWT P
573 removal are in their infancy, water and environmental managers will need to engage stakeholders
574 from across these disciplines at an early stage of development. This will ensure that strategies for
575 managing P sources to the environment consider both WWT-P and MWL-P, and that reductions in
576 WWT-P are not offset by increases in MWL-P. Where these practices are already well established,
577 integration of MWL-P into leakage targets and catchment P permits may be an effective policy
578 intervention (Goody et al. 2017). Adopting these strategies will ensure that P sources to the
579 environment are managed effectively, whilst safeguard human health.

580

581 **4.3 Research priorities for MWL-P**

582

583 Based on the uncertainties highlighted above, there are a number of key research priorities which
584 need to be addressed if MWL-P and its associated seasonality are to be effectively integrated into P
585 management strategies. The fate of water following a leak is currently poorly understood. It can be
586 hypothesised that the fate will vary depending on the type of leak as follows: (1) Visible winter bursts
587 causing rapid transport to rivers, groundwater and sewers, (2) Invisible winter leakage with a greater
588 recharge to groundwater, (3) Summer leakage associated with changes in soil moisture deficit (SMD)
589 causing possible loss by evapotranspiration and to groundwater, (4) Background leakage with a slower
590 transport and greater recharge to groundwater. Background leakage is likely to have the largest
591 impact in the long term, because these leaks are relatively difficult to locate and stop relative to bursts
592 (Edie 2016, Lambert 1994).

593

594 The fate of P within the subsurface following a leak adds further uncertainty. The pH of mains water
595 is often increased compared to raw untreated waters (Flem et al. 2015) which will inhibit formation
596 of iron and aluminium phosphate precipitates, resulting in increased P mobility. However, after
597 release into the environment MWL-P has a wide range of potential fates (clay sorption, soil/sediment
598 accumulation, flow to rivers, groundwater or the sewer network). Depending on the time of year and
599 the type of leak identified above, the fate of MWL-P may vary substantially. Visible winter bursts and
600 invisible winter leakage not captured by the sewer network may result in rapid P transport to rivers
601 and groundwater, potentially accelerated by high rainfall events following cold weather. Summer
602 leakage associated with changes in soil moisture and background leakage is likely to result in slow
603 transport and potentially sorption and accumulation of P in soils. Understanding the relative
604 significance of these different fates and lags in the hydrological system is essential if MWL-P is to be
605 managed in the future.

606

607 Long term changes in MWL-P seasonality are also likely to occur due to climate change. Climate
608 projections (UKCP09 2080s, Medium emissions scenario) show increases in daily mean, maximum and
609 minimum temperatures across the UK, with slightly greater increases in summer than winter. Whilst
610 changes to annual precipitation total are predicted to be small, winter and summer precipitation are
611 predicted to increase by up to 33% and decrease by up to 40% respectively (Met Office 2010, Murphy
612 et al. 2009). Warmer winters may reduce winter bursts, and warmer summers may increase leakage
613 related to soil movement. Consequently it is plausible that the seasonal component of MWL-P may
614 become more bimodal, with peaks in both winter and summer. Differences in the temporal rainfall
615 distribution are also likely to impact the fate of P within the wider catchment. Increased winter rainfall
616 may result in more pronounced winter P flushing (Johnson et al. 2009). Interactions between climate
617 change induced temperature and rainfall effects on P fluxes are likely to be complex and require
618 further investigation.

619

620

621

622 **5 Conclusions**

623

624 This study has quantified seasonality and future scenarios of fluxes of P from mains water leakage
625 (MWL-P) to the environment for the first time. MWL-P shows a strong seasonality, with peak fluxes
626 during burst and leak events in winter > 150% of fluxes during other seasons. During peak events,
627 MWL-P is equivalent to c. 20% of P fluxes from wastewater treatment works (WWT-P). A moderate
628 cross correlation between WWT-P and riverine P fluxes is observed as the short term temporal
629 variations in both of these fluxes are the result of winter rainfall events. This masks any potential

630 correlation between MWL-P and riverine P fluxes. A substantial increase in the ratio of MWL-P to
631 WWT-P is predicted to 2040 associated with implementation of substantial wastewater P removal and
632 minimal mains water leakage reduction. Further research is required to understand the fate of MWL-
633 P in the environment, future changes in MWL-P loadings, and potential approaches to integrate this P
634 source into water management strategies.

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636

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640

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