Ascott, M.J.; Gooddy, D.C.; Lapworth, D.J.; Davidson, P.; Bowes, M.J.; Jarvie, H.P.; Surridge, B.W.J. 2018. **Phosphorus fluxes to the environment from mains water leakage: seasonality and future scenarios.** *Science of the Total Environment*, 636. 1321-1332. https://doi.org/10.1016/j.scitotenv.2018.04.226

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

Ascott, M.J.[a], Gooddy, D.C.[a], Lapworth, D.J.[a], Davidson, P.[b], Bowes, M.J.[c], Jarvie, H.P.[c] and Surridge, B.W.J.[d]

[a] British Geological Survey, Maclean Building, Crowmarsh, Oxfordshire, OX10 8BB, United Kingdom.

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

[c] Centre for Ecology & Hydrology, Maclean Building, Crowmarsh, Oxfordshire, OX10 8BB, United Kingdom.

[d] Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom.

Corresponding Author: Ascott, M.J.*

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

Abstract

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

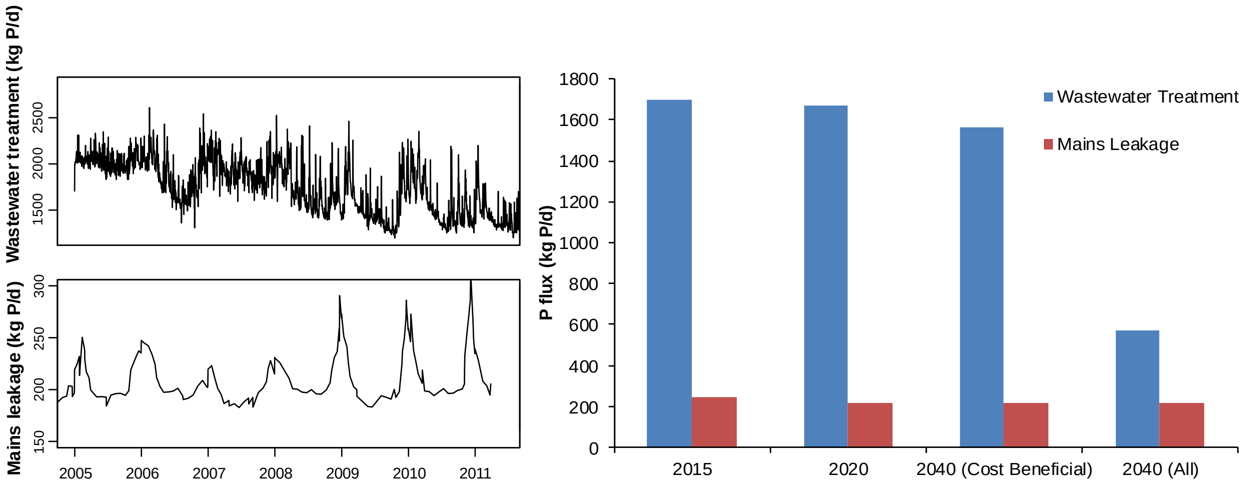
Keywords

Phosphorus, source apportionment, eutrophication, mains water, leakage

Highlights

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

Graphical Abstract



1 Introduction

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

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

Management 2011). In the USA, over half of supplies are dosed (Dodrill and Edwards 1995). Dosing achieves final P concentrations in tap water that are estimated to be between 700 – 1900 µg P/L (UK Water Industry Research Ltd 2012), with phosphate dosing having been a highly successful engineering solution to reduce lead concentrations in drinking water. For example, in 2011, 99.0% of random tap water samples in England and Wales met the drinking water limit of 10 µg Pb/L (Chartered Institute for Water and Environmental Management 2011).

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

However, understanding the temporal variability of MWL-P is also important, because biological impacts of P in aquatic ecosystems also show significant temporal variability, with autotrophic biomass growth occurring in many systems primarily during spring (March, April, May) and early summer (June, July, August) (Bowes et al. 2012a). A number of studies (Birek et al. 2014, Cocks and Oakes 2011, UK Water Industry Research Ltd 2007, 2013) have highlighted seasonal dynamics in leakage of water from mains supplies. Higher rates of both bursts and low-level continuous "invisible" leakage occur during winter (December, January, February), due to pipe contraction and expansion. However, work on P to date has only quantified the annual flux of P from mains water leakage and the temporal dynamics of this source are poorly constrained. Whilst substantial reductions in WWT-P loads to rivers have been achieved (Worrall et al. 2015), elevated P concentrations remain a significant concern, with 40% of the water bodies in England not achieving "good status" under the Water Framework Directive (European Union 2000) due to high reactive P concentrations (Environment Agency 2015). Consequently, in England and Wales, substantial further investment in wastewater treatment is planned to 2020 with an estimated total expenditure of £2.1 billion (Global Water Intel 2014). Similar programmes exist internationally, for example in the USA (Sewage World 2016) and across Europe (Water Technology 2016). In parallel, water companies continue to reduce leakage based on water saving drivers, which will in turn reduce MWL-P fluxes. Despite these programmes, no estimates of how the relative contributions of MWL-P and WWT-P to the environment may change in the future have been made. If P sources to aquatic ecosystems are to be managed effectively, and the impacts of MWL-P reduced, it is essential that the temporal dynamics of MWL-P and potential future changes in fluxes from this source of P are better constrained. The research reported here was undertaken to examine the following hypotheses:

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

Using the River Thames, a large lowland catchment in the UK, as an exemplar, we tested these hypotheses by deriving historic, daily MWL-P, WWT-P and riverine P fluxes. Subsequently we validated this novel approach by comparing our derived annual fluxes of P from MWL and WWT sources to fluxes reported in previous studies. We then undertook time series analysis using multiple linear regression modelling to develop an improved understanding of the temporal dynamics and the processes controlling MWL-P, WWT-P and riverine P fluxes. Further, we derived future scenarios for MWL-P and WWT-P, based on 25 year plans for leakage reduction and WWT-P removal respectively as published by the UK environmental regulator and water companies. Finally, we provide a summary of key research priorities in the context of MWL-P, and consider how to manage this source of P across a range of different hydro-socioeconomic settings.

2 Materials and Methods

2.1 Study area

We quantified daily MWL-P, WWT-P and riverine P fluxes for the non-tidal Thames catchment (Figure 1). The Thames is a relatively large (9948 km^2) lowland catchment in southern England. The catchment is predominantly underlain by the Cretaceous Chalk, with Oolitic limestones at the head of the catchment and areas of low permeability clays around Oxford and London. Mean annual rainfall in the catchment at Oxford is 745 mm (Marsh and Hannaford 2008). Although the catchment is relatively rural, with approximately 45% of land classified as arable (Fuller et al. 2013), there are also a number of major urban areas (London, Oxford, Reading) resulting in a high catchment population density (960 people/km^2 (Merrett 2007) and relatively small agricultural P losses (<10% of all total phosphorus sources (White and Hammond 2009)).

Water supply in the catchment is provided by four different water companies (Thames Water, Affinity Water, Sutton and East Surrey Water and South East Water) which operate across a total of 10 largely self-contained water resource zones (WRZs, Environment Agency (2012)). Each individual water company reports annual leakage rates. Leakage rates are reported to be high, reaching up to 26% of water input to the distribution network (Committee on Climate Change 2015). The catchment has been subject to previous research to estimate the annual contribution of MWL-P and WWT-P to the River Thames (Gooddy et al. 2017). Building on this initial research, Ascott et al. (2016a) estimated that approximately 30% of the MWL-P flux in England and Wales was derived from the river basin district in which the non-tidal Thames catchment is located. Wastewater treatment occurs throughout the catchment and 137 water company wastewater treatment works serving > 2000 population equivalents (p.e) are present. Since the late 1990s, P fluxes from WWTs have been reduced in the catchment through the introduction of tertiary treatment, primarily driven by the EU Urban Waste Water Treatment Directive (Kinniburgh and Barnett 2010, Powers et al. 2016). Consequently, P concentrations in the River Thames fell by approximately 90% between 1992 and 2005 (Kinniburgh and Barnett 2010). Despite this, there is scant evidence that eutrophication risk in the Thames has reduced (Bowes et al. 2014), with a number of rivers in the catchment exhibiting excessive phytoplankton biomass and P concentrations above those at which the growth of autotrophs can be expected (> 80 μg SRP/l, Bowes et al. (2012b)).

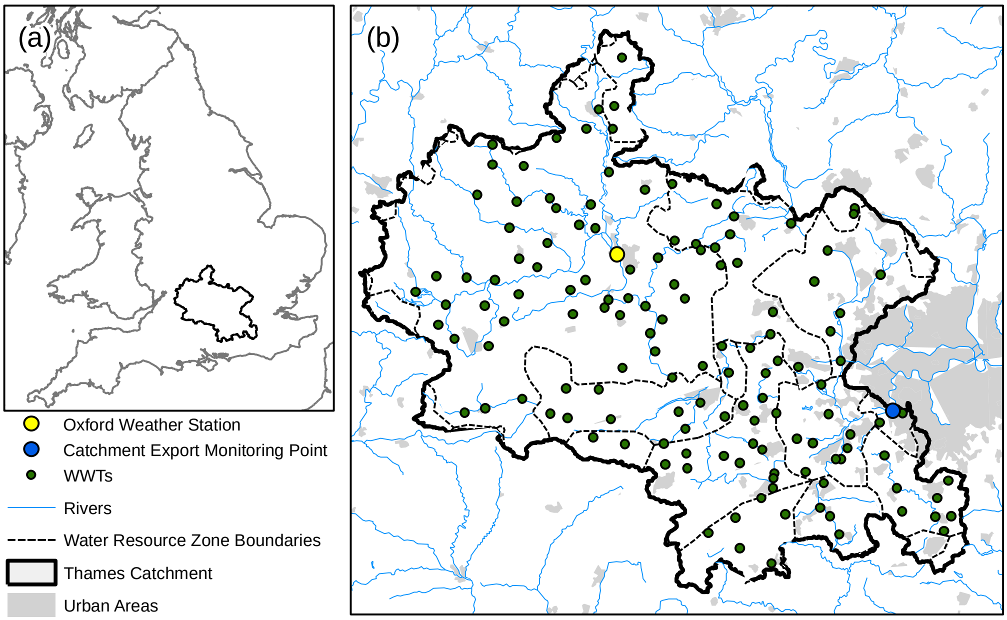


Figure 1 Location of the non-tidal Thames catchment within England (a) and the water resource zones, wastewater treatment works serving > 2000 p.e., catchment export monitoring point and the Oxford weather station within the catchment (b). Contains Ordnance Survey data © Crown copyright and database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

2.2 P flux time series derivation and analysis

2.2.1 Data sources

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

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	Units	Source
MWL-P	WRZ level leakage rates for WRZs in non-tidal Thames catchment	2011 - 2013	Annual	ML/day	WRMP tables
	Leakage rates for Thames Water	2001 - 2011	Daily	ML/day	Cocks and Oakes (2011)
	PO_4-P dosing concentration	2001 - 2011	Annual	mg P/L	Comber et al. (2011)
	PO_4-P dosing extent	2001 - 2011	Annual	-	Hayes et al. (2008), Chartered Institute for Water and Environmental Management (2011)
Riverine P	PO_4-P concentration for Thames at Teddington	2000 - 2015	Monthly	mg P/L	Environment Agency
	Flows for the Thames at Teddington	2000 - 2015	Daily	m^3/s	
WWT-P	PO_4-P WWT discharge concentrations	2005 - 2011	Variable (mean frequency = 10 days, median frequency = 27 days)	mg P/L	Environment Agency
	WWT discharge flows	2005 - 2011	Daily	m^3/s	

2.2.2 MWL-P

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

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

Riverine P flux derivation

The daily net export of P (2001-2016) from the catchment was derived using observed orthophosphate-P (PO_4-P) concentration and daily flow data for the River Thames at Teddington

(Figure 1). In order to infill missing concentration data to derive a daily P flux time series, we first undertook single change point detection using an asymptotic penalty value of 0.05 (Chen and Gupta 2011, Killick and Eckley 2014) to determine whether there was a statistically significant change in the mean and variance of the concentration time series associated with historic reductions in P loads from WWTs (Kinniburgh and Barnett 2010). Within each of the datasets divided by the change point, we examined whether concentration-flow relationships also change associated with season and flow conditions ((Moatar et al. 2017, Zhang 2018, Zhang et al. 2016)). No significant differences were found under different season and flow conditions, and so single concentration-flow relationships before and after any change points were used for each following the non-linear least-squares regression approach of Bowes et al. (2014):

$$C = AQ^{B} \tag{1}$$

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

Wastewater Treatment Works P Flux Estimation

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

For the monitored WWT sites, daily flow monitoring data for 2005 – 2011 were used in conjunction with PO_4-P concentration data to derive daily P fluxes for each site. A similar approach to that described above for the riverine P flux estimates was used for infilling of missing concentration data using concentration-flow relationships of the form of equation 1. The dates for implementation of P removal schemes at WWTs in the Thames catchment were provided by the environmental regulator for England. Where P removal schemes were implemented at a WWT during the study period, separate concentration-flow relationships were estimated for before and after implementation. Where no changes in treatment processes for P removal at WWTs occurred during the study period, a single concentration-flow relationship for the WWT was used. The derived daily P fluxes for each individual WWT were then summed to derive a total, daily catchment flux of WWT-P.

Flux comparison and time series analysis

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

In order to assess the control of cold weather events on temporal variability in MWL-P, we undertook correlation analyses using historic daily minimum temperature data for the Oxford station in the Thames catchment (Figure 1). We also quantified correlations between daily rainfall and daily WWT-P and daily riverine P loads to assess the role of short term rainfall events in controlling P fluxes. Finally, to determine how much of the variation in daily riverine P load could be explained by daily MWL-P and daily WWT-P, we developed parsimoniuous multiple linear regression models using (1) WWT-P only and (2) WWT-P + MWL-P as explanatory variables. To determine the impact of adding the MWL-P variable on model predictive power we initially compared the model results by visual inspection. We then compared the coefficient of determination between the two models and finally used a partial F-test (R Development Core Team 2016). There is substantial uncertainty in the fate of MWL-P in the environment associated with the relative importance of different P retention (clay sorption, soil and sediment accumulation) and transport processes (groundwater, surface water and the sewer network) (Gooddy et al. 2017). In this context, our research does not aim to make a direct causal link between time series variability in WWT-P, MWL-P and riverine P fluxes. The purpose of the comparison of fluxes described here is threefold: (1) to provide indications of the relative contributions of these sources to the environment; (2) to improve understanding of the processes controlling temporal variability and seasonality in these sources; and (3) to improve understanding of the relationships between the sources.

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

Scenarios for future reductions in both MWL-P and WWT-P were constructed on the basis of existing water company and environmental regulator investment plans. Planned leakage reductions for each of the 10 WRZs in the Thames catchment for 2015-2040 were extracted from water company water resource planning tables (Affinity Water 2014, Southeast Water 2014, Sutton and East Surrey Water 2014, Thames Water 2014). Estimates of future reductions in WWT-P were provided by the environmental regulator as new discharge permit limits for P (in mg P/L). In England and Wales investments in wastewater treatment are planned on a five-year Asset Management Programme (AMP) cycle. Investments over 2015 – 2020 (Asset Management Programme 6 (AMP6)) are fully planned and costed. Further into the future, a provisional "long list" of improvements in WWTs that are potentially required in order to meet good ecological status in the catchment has been derived based on source apportionment modelling (Comber et al. 2013). This list has been subject to a cost benefit analysis which has divided sites into those at which enhanced P removal is cost beneficial and those at which this is not. In total, four WWT improvement scenarios were developed, as detailed below:

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

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

3 Results

3.1 MWL-P flux estimation

Figure 2 shows the evolution of the MWL-P flux over the period 2001 to 2011 for the Thames catchment. Figure 2 (c) shows the total daily leakage (Cocks and Oakes 2011) for 2001 – 2011 for Thames Water. Over the period 2001 – 2011, the volume of leakage has declined due to water companies actively locating and fixing leaks in the distribution network. After accounting for this reduction, the leakage time series shows a relatively strong correlation at lag = 0 with daily minimum temperature (Pearson correlation, $r = 0.63$, $p < 0.001$). The time series shows a clear seasonal signal associated with increased bursts and invisible leakage in winter . The winters of 2009, 2010 and 2011 were particularly cold, with mean December temperatures 1.1, 2.0 and 4.8 degrees below long term average (1971-2000) respectively (Met Office 2016). This resulted in large increases in leakage of up to 50% compared to the preceding autumn months (September, October, November). In summer (June, July, August), small increases of up to 6% compared to spring (March, April, May) values are also observed, which has been attributed to soil expansion and contraction resulting in pipe failure (Cocks and Oakes 2011). Figure 2 (d) shows the estimated daily flux of MWL-P for 2001-2011 for the Thames catchment. Whilst the volume of leakage has reduced substantially, increases in the spatial extent of dosing and in phosphate dosing concentrations counteract these reductions, resulting in significant (Mann Kendall trend test, $p < 0.001$) net increases in MWL-P fluxes of 300% over the period 2001 to 2011. Seasonality increases through time with a significant change in variance identified at 08/09/2008 (single change point detection using an asymptotic penalty value = 0.01) and the ratio of daily annual maximum/minimum MWL-P rates increasing from 130% in 2002 – 2008 to 150% in 2008 – 2011.

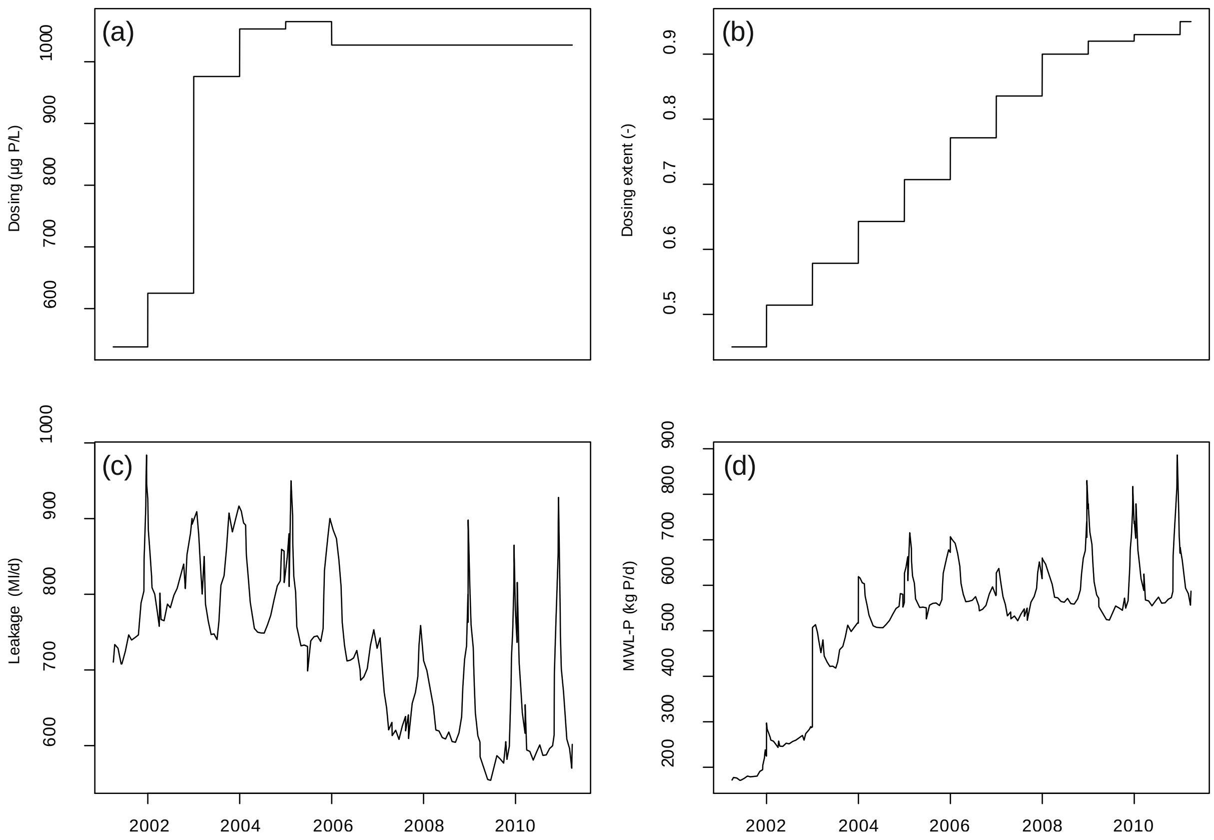


Figure 2: Estimates of P dosing concentrations (a) and extents (b) for 2001 – 2011, daily leakage rates reported by Cocks and Oakes (2011) (c), and the derived MWL-P flux (d)

3.2 WWT-P flux estimation

Figure 3 reports data related to the WWT-P flux for the Thames catchment. Substantial decreases in P concentration in WWT effluents occurred between 2005 and 2007, associated with the introduction of P removal technology. From March 2008 to 2012, no significant further P removal was implemented in the catchment. Consequently, effluent P concentrations have been relatively stable and the frequency of P monitoring reduced. The slight increase (5.4 to 5.6 mg P/L) in effluent P concentrations over this period may reflect an overall decrease in P removal efficiency in existing

WWTs due to changes in plant operation and influent P loads (Tchobanoglous and Burton 2013). There is considerable variation in the WWT flows (Figure 3 (a)) particularly during high flow periods in winter associated with rainfall events when flows can be up to twice as large as dry weather flows (DWF). The derived WWT-P flux (Figure 3 (c)) reflects both long term changes in P concentrations following investment in P removal technologies and short term variability in WWT flows associated with rainfall events.

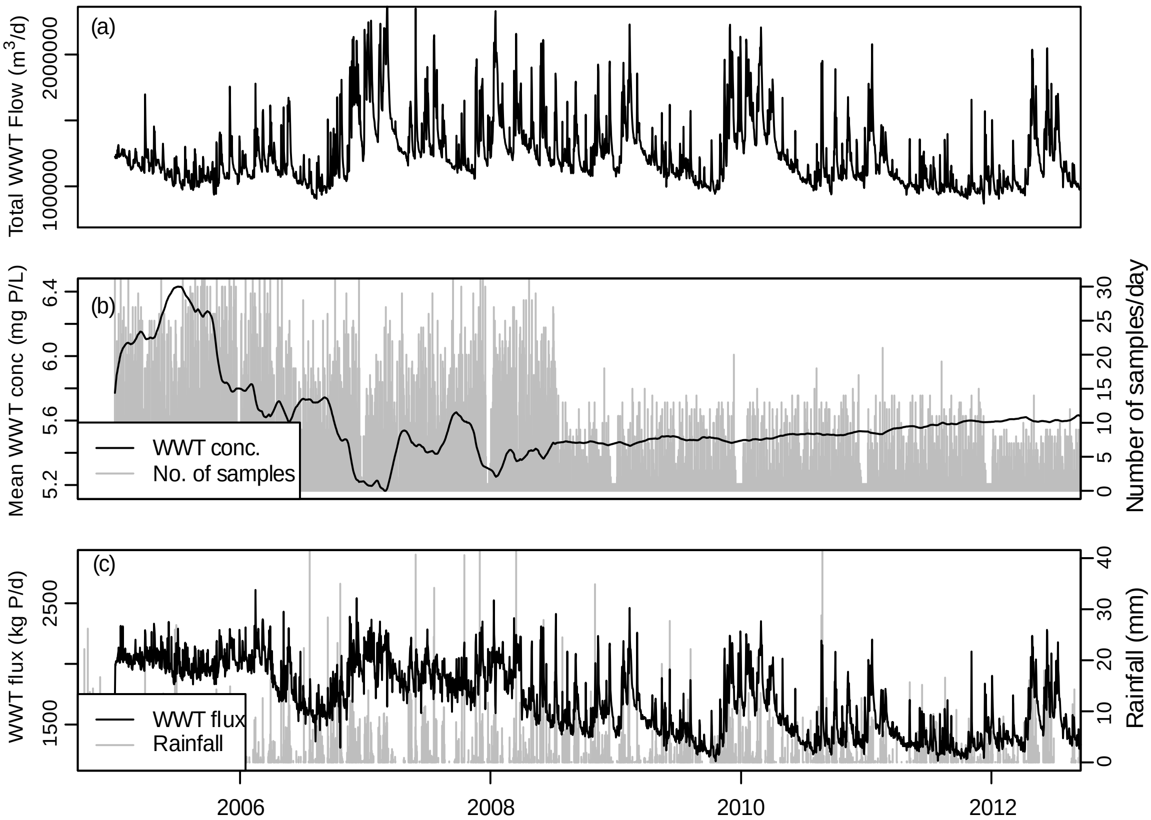


Figure 3 Total WWT flows (a), mean daily P concentrations and number of samples (b) and the derived WWT-P flux and rainfall time series for Oxford (c)

3.3 Flux comparison

Table 2 reports the annual mean MWL-P and WWT-P fluxes derived in the research reported here and in previous studies of the Thames catchment. The annual estimates of MWL-P in this study are within the range of values reported by Gooddy et al. (2017) who considered a range of different plausible historic mains water dosing extents. WWT-P fluxes broadly corroborate previous findings reported by Haygarth et al. (2014) and Comber et al. (2013). Fluxes estimated in the current study for 2006-2008 are somewhat higher (29%) than estimates made using observed soluble reactive P concentrations in WWT discharges in conjunction with source apportionment tools (Comber et al. 2013). Fluxes from the source apportionment modelling were adjusted by the UK environmental regulator (Environment Agency 2016) to take into account new and enhanced P removal (and hence lower WWT-P fluxes) which is the likely cause of this discrepancy. Our estimates are also larger (50%) than those calculated by Haygarth et al. (2014), who used a simple approach based on population equivalents and assumed total P concentrations to estimate WWT-P flux for the catchment.

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

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

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

3.4.1 Riverine P flux estimation

Figure 4 shows the results of the change point detection analysis and the derived concentration-flow relationships before and after the change point for the River Thames at Teddington. A substantial decrease in the mean and variance of riverine phosphate concentrations was observed in 2006, associated with reductions in P loads from WWTs. For the period 2000 – 2006, the concentration-flow relationship shows decreases in concentrations with increasing flow, associated with dilution of WWT inputs. This corroborates concentration flow relationships derived by Bowes et al. (2014) in the Thames basin. From 2006 to 2015, the variability in concentration with flow is substantially reduced. Between 0 and 100 m^3/d decreases in concentration with increasing flow are observed, associated with dilution of point source P inputs. However, from 100 to 500 m^3/d, there is limited change in concentration with increasing flow which reflects a balance of both point source dilution and mobilisation of diffuse P sources across this range of discharges.

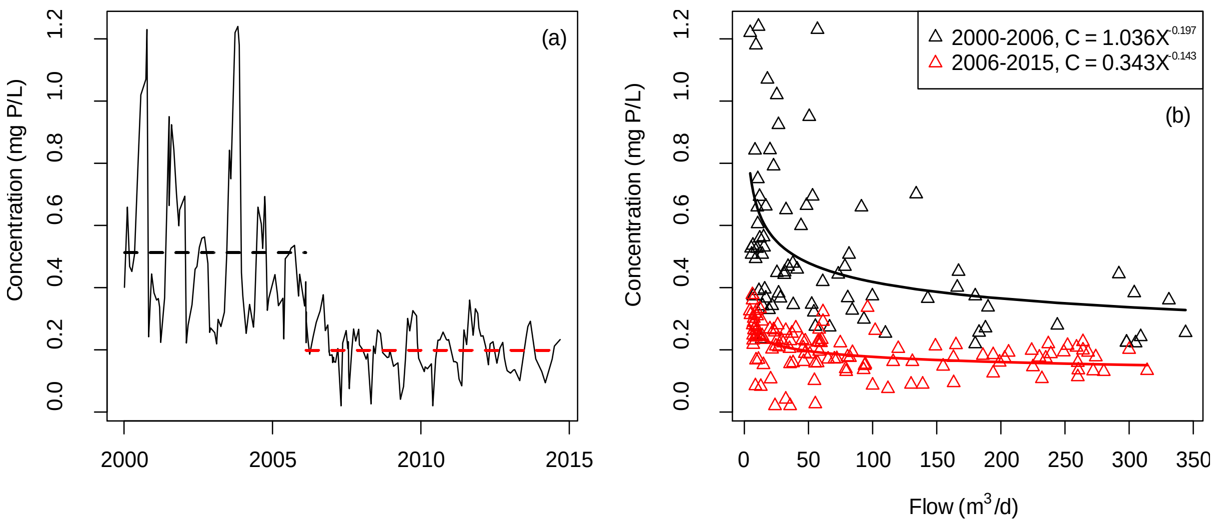


Figure 4 Observed phosphate concentrations (mg P/L) in the River Thames at Teddington and mean concentrations (dashed line) before and after the change point (a) and concentration-flow relationships for each of these periods (b).

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

Figure 5 shows the ratio of MWL-P and WWT-P fluxes ("MWL-P/WWT-P") and their relationship with P flux in the River Thames at Teddington. From 2005 to 2011 there is a significant trend in MWL-P/WWT-P (Mann-Kendall trend test, $p < 0.001$). The ratio of MWL-P to WWT-P increases from from 7-10 in 2005 to 15% in 2011. This is due to two factors: (1) an increase in the extent of mains water

P dosing through the period, as shown in Figure 2; and (2) a decrease in WWT-P loads due to investment in P removal technology.

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

Figure 5 (a) and (d) show the daily WWT-P flux and catchment P export. There are weak but significant correlations between WWT-P and daily rainfall (Pearson correlation, $r = 0.23$, $p < 0.001$) and catchment export and daily rainfall (Pearson correlation, $r = 0.18$, $p < 0.001$). Figure 6 and Figure 7 show observed and modelled riverine P fluxes using MLR models considering: (1) WWT-P only; and (2) WWT-P and MWL-P together. Figure 8 shows modelled and observed riverine P fluxes in detail for winter months from 2005 to 2011. Adding MWL-P as a predictor variable increases the modelled riverine P flux associated with increased MWL-P fluxes in winter. This is particularly the case in the winters of 2008/9, 2009/10 and 2010/11 where increases in winter leakage were particularly large (Figure 2). The multiple linear regression model used to determine whether the riverine P load can be explained by MWL-P and WWT-P resulted in a relatively modest coefficient of determination (adjusted $R^2 = 0.41$). The MLR model suggested that WWT-P and MWL-P are both significant explanatory variables ($p < 0.001$). Removing MWL-P from the regression significantly reduced the predictive power of the model ($F = 242$, $p < 0.001$, for partial F-test between MLR models considering WWT-P only and WWT-P + MWL-P) resulting in a lower coefficient of determination (adjusted $R^2 = 0.33$).

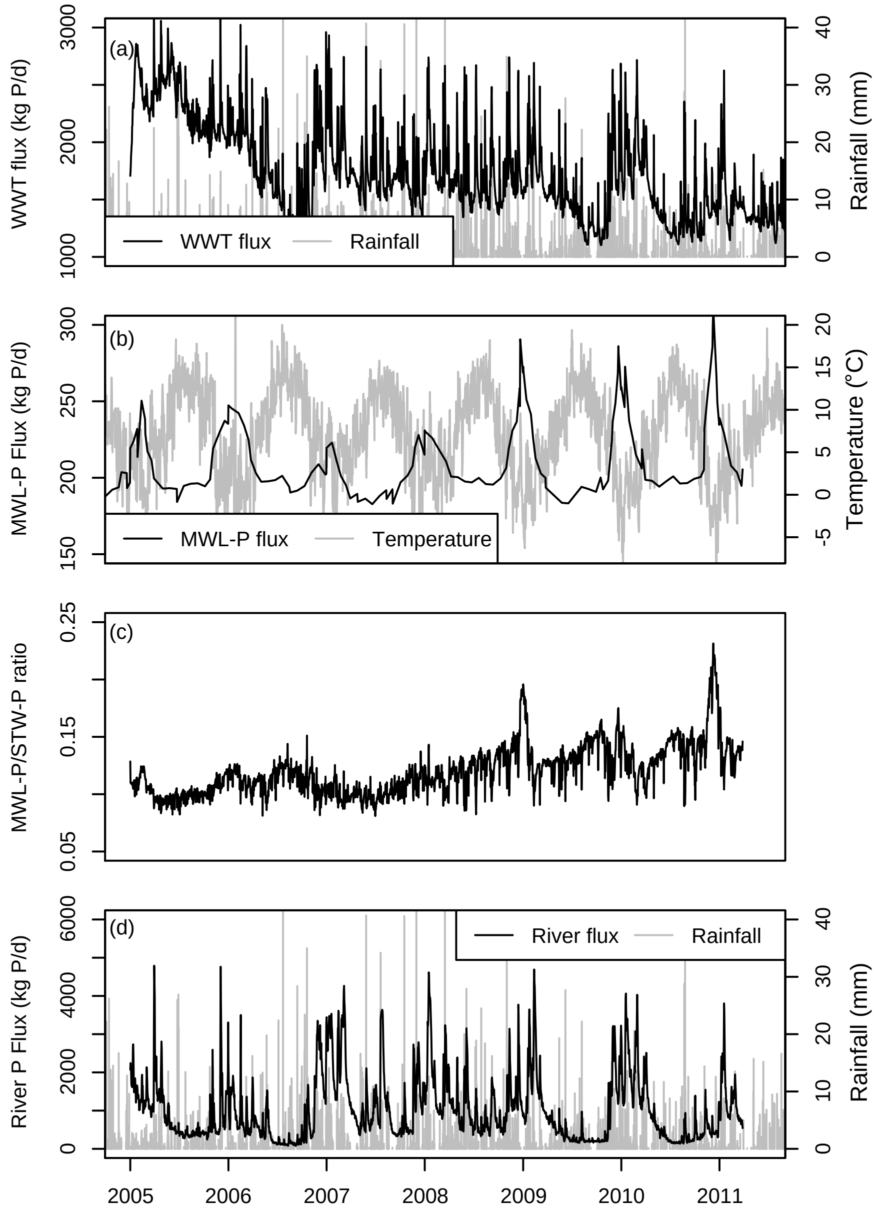


Figure 5 Daily fluxes of WWT-P and rainfall (a), MWL-P and temperature (b) for the Thames catchment, the ratio of MWL-P to WWT-P (c) and the riverine flux of P out of the catchment and rainfall (d)

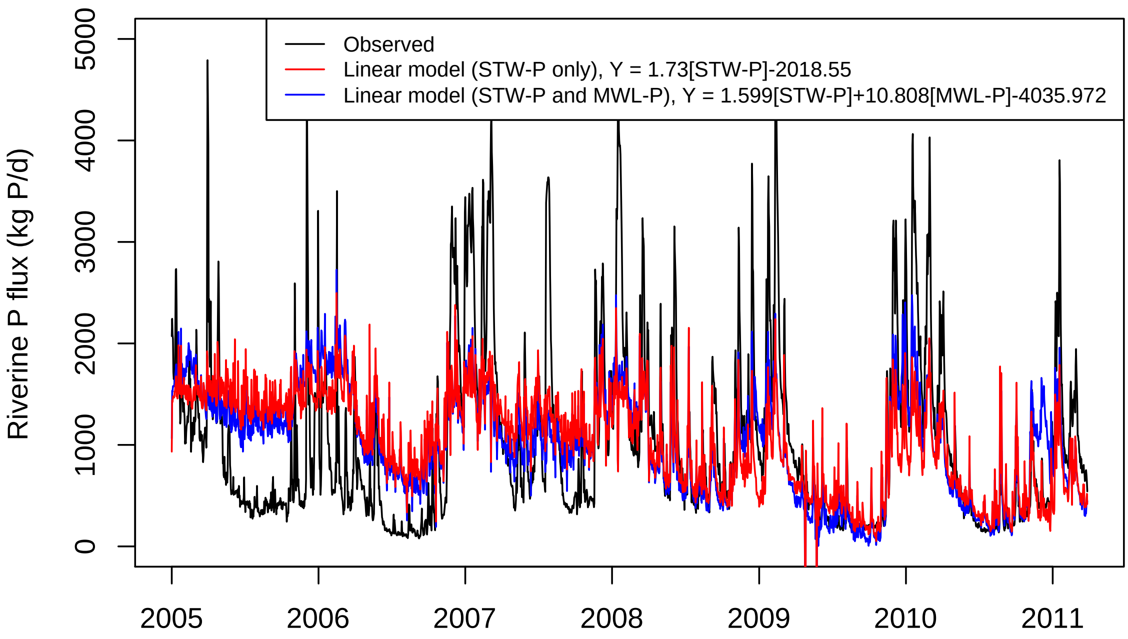


Figure 6: Observed (black) and modelled riverine P fluxes for the export of the Thames catchment using the MLR model considering WWT-P only (blue) and WWT-P and MWL-P (red)

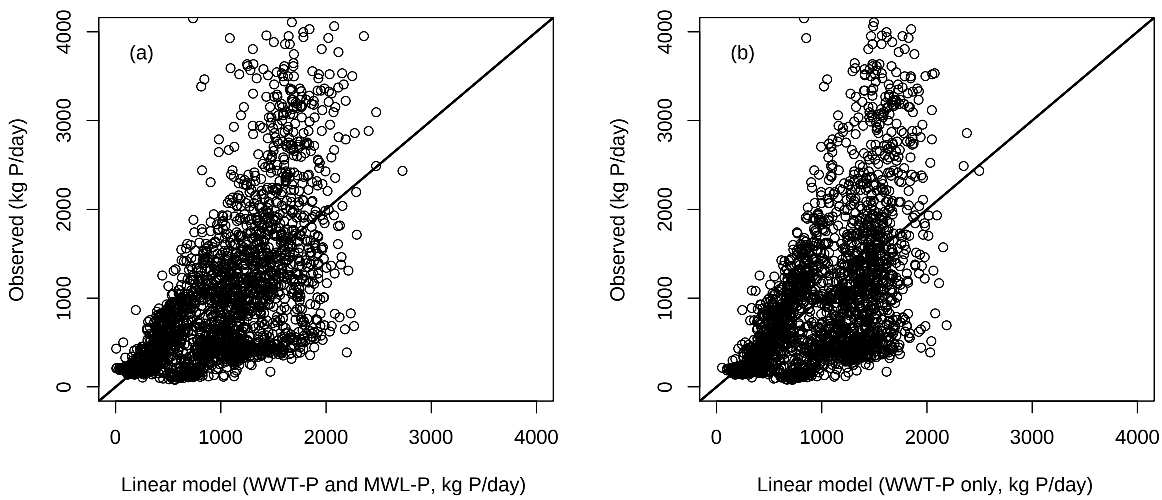


Figure 7 Observed and modelled riverine P fluxes for linear model considering WWT-P and MWL-P (a) and WWT-P only (b)

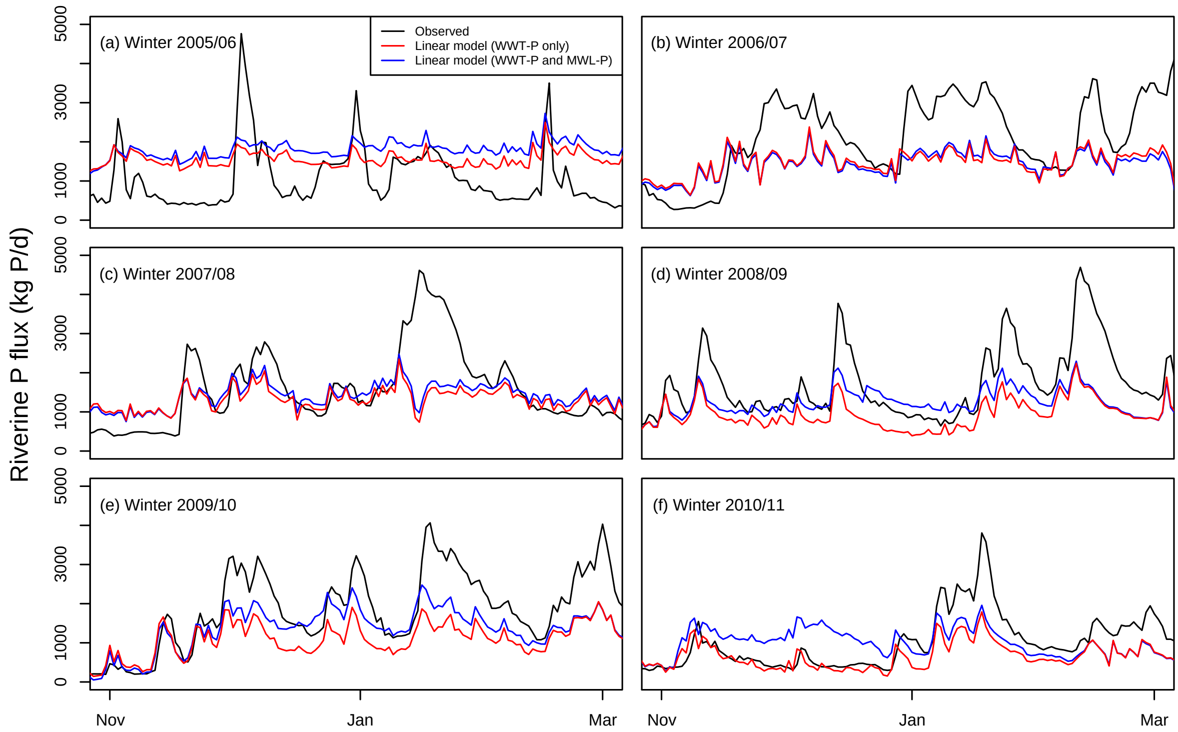


Figure 8 Riverine P flux time series for winter months for 2005/06 (a) to 2010/12 (f)

3.5 Future loads of MWL-P and WWT-P

Table 3 reports the derived future MWL-P and WWT-P loads for annual average conditions. WWT-P fluxes are predicted to decrease to 2020 associated with the AMP6 programme, with further decreases predicted to 2040. When considering all WWT improvements to 2040, a total decrease in 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 occur to 2040. In total, the reduction in MWL-P loading is approximately 2% of the reduction in WWT-P. Consequently, the ratio of MWL-P to WWT-P is predicted to increase from 15% to 38% under average conditions.

Table 3 Future MWL-P and WWT-P loadings

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

4 Discussion

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

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

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

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

In the context of MWL-P, there is some uncertainty in the extent of P dosing and the concentrations used. In this study, we used a conservative approach to estimate historic changes in P dosing concentrations and extents. It is plausible, however, that dosing extents and concentrations may have been higher than estimated here, which would result in greater flux seasonality. Just as water companies publically release water resource planning data, making historic P concentrations and dosing extents available would be beneficial to refining MWL-P estimates. Given the potential significance of this flux, assessment of MWL-P using observed leakage and P concentration data at the local scale using district metered area (DMA) data could provide important new insights. Further, the uncertainty surrounding the ultimate fate of MWL-P following leakage is significant. As discussed previously, comparisons with WWT-P and riverine P fluxes such as those presented in Figure 5 are beneficial as they provide indications of the relative contribution of different sources to the environment. It is likely that MWL-P makes some direct contribution to riverine P fluxes, but the relative importance of groundwater, surface water and the sewer network as pathways for MWLP-P are largely unknown at this time. Moreover, the spring and summer growing period is often perceived to be the most biologically-critical period in many receiving waters (Jarvie et al. 2006), meaning that winter peaks in MWL-P which do directly contribute to riverine P loads may have a limited immediate impact on riverine ecosystems. However, there are likely to be time lags between the flux of P from a water mains leak and this flux reaching a river, with a range of transit times depending on catchment size, transport pathway and change in P concentration along the pathway. Consequently, it is plausible that winter peaks in leakage of P from water mains only reach a river network in more biologically-critical periods of the year. Long transit times are likely in catchments with a significantly thick unsaturated zone and long groundwater flow paths. In these catchments it is plausible that MWL-P contributes to legacy P stores in groundwater (Holman et al. 2008) as has been observed for nitrate (Ascott et al. 2017, Ascott et al. 2016b). River sediments can also accumulate P (Sharpley et al. 2013), with time lags of < 1 year associated with in-stream P remobilisation during high river flow events. These in-stream accumulation-remobilisation processes will result in further lags between MWL-P that reaches a river and P riverine exports. Whilst not considered in this study, agriculture remains a significant P source to the environment in many catchments (White and Hammond 2009) and should clearly be accounted for in future P management strategies. Much of the likely temporal dynamics of the fate of MWL-P (higher winter fluxes with potential catchment legacy stores and lags) are analogous to the fate of nonpoint P sources such as agriculture (Sharpley 2016). Improved understanding of the fate of these P sources could be developed through high frequency riverine P monitoring and application of data driven modelling tools (e.g Chen and Chau (2016), Olyaie et al. (2015), Taormina et al. (2015), Wang et al. (2014)). The anticipated impact of measures to reduce P fluxes from both mains leakage and agriculture should be tempered with knowledge that catchment lags and release of existing nonpoint legacy P sources may significantly delay any water quality improvements.

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

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

A number of European countries have adopted policies that actively avoid phosphate dosing on environmental grounds (Chartered Institute for Water and Environmental Management 2011). Countries such as the Netherlands undertake pH correction and centralized water softening to ensure lead concentrations are below the required standard (Hayes 2012). P has been shown to be a limiting nutrient to biofilm growth in a number of different drinking water systems (Liu et al. (2016) and references therein) and concern has been raised historically that the presence of P in water mains (through dosing or otherwise) would result in increased bacterial counts in water mains (Miettinen et al. 1996). The current and future significance of MWL-P in comparison to other P sources identified in this study adds to the body of evidence to support policies that avoid phosphate dosing, where the extent of lead piping in the distribution network is small enough to safeguard human health protected without dosing.

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

4.3 Research priorities for MWL-P

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

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

Changes in seasonality of P fluxes have not been quantified in this study and should be considered in future research. Long term changes in intra-annual variability in MWL-P are likely to occur due to climate change. Climate projections (UKCP09 2080s, Medium emissions scenario) show increases in daily mean, maximum and minimum temperatures across the UK, with slightly greater increases in summer than winter. Whilst changes to annual precipitation total are predicted to be small, winter and summer precipitation are predicted to increase by up to 33% and decrease by up to 40% respectively (Met Office 2010, Murphy et al. 2009). Warmer winters may reduce winter bursts, and warmer summers may increase leakage related to soil movement. Consequently it is plausible that the seasonal component of MWL-P may become more bimodal, with peaks in both winter and summer. Differences in the temporal rainfall distribution are also likely to impact the fate of P within the wider catchment. Increased winter rainfall may result in more pronounced winter P flushing (Johnson et al. 2009). Interactions between climate change induced temperature and rainfall effects on P fluxes are likely to be complex and require further investigation.

5 Conclusions

This study has quantified seasonality and future scenarios of fluxes of P from mains water leakage (MWL-P) to the environment for the first time. MWL-P shows a strong seasonality, with peak fluxes during burst and leak events in winter > 150% of fluxes during other seasons. During peak events, MWL-P is equivalent to approximately 20% of P fluxes from wastewater treatment works (WWT-P). A moderate cross correlation between WWT-P and riverine P fluxes is observed as the short term temporal variations in both of these fluxes are the result of winter rainfall events. This masks any potential correlation between MWL-P and riverine P fluxes. A substantial increase in the ratio of MWL-P to WWT-P is predicted to 2040 associated with implementation of substantial wastewater P removal and minimal mains water leakage reduction. Further research is required to understand the fate of MWL-P in the environment, future changes in MWL-P loadings, and potential approaches to integrate this P source into water management strategies.

6 Acknowledgements

The research was funded by the UKs Natural Environment Research Council (NERC) National Capability resources devolved to the British Geological Survey. This paper is published with permission of the Executive Director, British Geological Survey (NERC).

References

Affinity Water. Latest Reports. https://stakeholder.affinitywater.co.uk/latest-reports.aspx. Hatfield, UK: Affinity Water, 2014.

Ascott, M.J., Gooddy, D.C., Lapworth, D.J. and Stuart, M.E. (2016a) Estimating the leakage contribution of phosphate dosed drinking water to environmental phosphorus pollution at the national-scale. Science of The Total Environment 572, 1534-1542.

Ascott, M.J., Gooddy, D.C., Wang, L., Stuart, M.E., Lewis, M.A., Ward, R.S. and Binley, A.M. (2017) Global patterns of nitrate storage in the vadose zone. Nature Communications 8(1), 1416.

Ascott, M.J., Wang, L., Stuart, M.E., Ward, R.S. and Hart, A. (2016b) Quantification of nitrate storage in the vadose (unsaturated) zone: a missing component of terrestrial N budgets. Hydrological Processes 30(12), 1903-1915.

Bellinger, D., Leviton, A., Waternaux, C., Needleman, H. and Rabinowitz, M. (1987) Longitudinal analyses of prenatal and postnatal lead exposure and early cognitive development. New England journal of medicine 316(17), 1037-1043.

Birek, L., Petrovic, D. and Boylan, J. (2014) Water leakage forecasting: the application of a modified fuzzy evolving algorithm. Applied Soft Computing 14, Part B, 305-315.
Bowes, M.J., Gozzard, E., Johnson, A.C., Scarlett, P.M., Roberts, C., Read, D.S., Armstrong, L.K., Harman, S.A. and Wickham, H.D. (2012a) Spatial and temporal changes in chlorophyll-a concentrations in the River Thames basin, UK: Are phosphorus concentrations beginning to limit phytoplankton biomass? Science of The Total Environment 426, 45-55.
Bowes, M.J., Ings, N.L., McCall, S.J., Warwick, A., Barrett, C., Wickham, H.D., Harman, S.A., Armstrong, L.K., Scarlett, P.M., Roberts, C., Lehmann, K. and Singer, A.C. (2012b) Nutrient and light limitation of periphyton in the River Thames: Implications for catchment management. Science of The Total Environment 434, 201-212.
Bowes, M.J., Jarvie, H.P., Naden, P.S., Old, G.H., Scarlett, P.M., Roberts, C., Armstrong, L.K., Harman, S.A., Wickham, H.D. and Collins, A.L. (2014) Identifying priorities for nutrient mitigation using river concentration–flow relationships: The Thames basin, UK. Journal of Hydrology 517, 1-12.
Bowes, M.J., Smith, J.T., Jarvie, H.P. and Neal, C. (2008) Modelling of phosphorus inputs to rivers from diffuse and point sources. Science of The Total Environment 395(2), 125-138.
Chartered Institute for Water and Environmental Management. Lead in Drinking Water. http://www.ciwem.org/policy-and-international/policy-position-statements/lead-in-drinking-water.aspx, 2011.
Chen, J. and Gupta, A.K. (2011) Parametric statistical change point analysis: with applications to genetics, medicine, and finance, Springer Science & Business Media.
Chen, X.Y. and Chau, K.W. (2016) A Hybrid Double Feedforward Neural Network for Suspended Sediment Load Estimation. Water Resources Management 30(7), 2179-2194.
Cocks, R. and Oakes, A. (2011) Modelling the Impact of Weather on Leakage and Bursts. Savic, D.A., Kapelan, Z. and Butler, D. (eds), pp. 351-357, University of Exeter, University of Exeter.
Comber, S., Cassé, F., Brown, B., Martin, J., Hillis, P. and Gardner, M. (2011) Phosphate treatment to reduce plumbosolvency of drinking water also reduces discharges of copper into environmental surface waters. Water and Environment Journal 25(2), 266-270.
Comber, S.D., Smith, R., Daldorph, P., Gardner, M.J., Constantino, C. and Ellor, B. (2013) Development of a chemical source apportionment decision support framework for catchment management. Environmental Science & Technology 47(17), 9824-9832.
Committee on Climate Change (2015) CCRA2: Updated projections of water availability for the UK.

De Bénédittis, J. and Bertrand-Krajewski, J.-L. (2005) Infiltration in sewer systems: comparison of measurement methods. Water Science and Technology 52(3), 219-227.

Deloitte (2014) Water Country Profiles, Deloitte Touche Tohmatsu Ltd, London.

Dodrill, D.M. and Edwards, M. (1995) Corrosion control on the basis of Utility Experience. Journal American Water Works Association 87(7), 74-85.

Dubrovsky, N.M., Burow, K.R., Clark, G.M., Gronberg, J., Hamilton, P.A., Hitt, K.J., Mueller, D.K., Munn, M.D., Nolan, B.T. and Puckett, L.J. (2010) The quality of our Nation's waters-Nutrients in the Nation's streams and groundwater, 1992-2004, US Geological Survey.

Edie. Leakage Explained. http://www.edie.net/abb/view_entry.asp?id=4268, 2016.

Edwards, M., Triantafyllidou, S. and Best, D. (2009) Elevated Blood Lead in Young Children Due to Lead-Contaminated Drinking Water: Washington, DC, 2001–2004. Environmental Science & Technology 43(5), 1618-1623.

Elser, J.J., Bracken, M.E.S., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., Ngai, J.T., Seabloom, E.W., Shurin, J.B. and Smith, J.E. (2007) Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. Ecology Letters 10(12), 1135-1142.

Environment Agency (2012) Water Resources Planning Guideline: The Technical Methods and Instructions, Environment Agency, Bristol, UK.

Environment Agency (2015) WFD Water Bodies: 2015 status and reasons for not achieving good (RNAG) for the update to the river basin management plans - Cycle 2.

Environment Agency (2016) Water company P loads discharged into environment in freshwater River Thames catchment, Environment Agency, Bristol, UK.

European Commision (1998) Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption, European Commision, Brussels.

European Union (1991) Council Directive of 21 May 1991 concerning urban waste water treatment (91/271/EEC), European Union, Brussels.

European Union (2000) Directive 2000/60/EC of the European Parliament and of the Council establishing a framework for the Community action in the field of water policy, European Union, Brussels.

Flem, B., Reimann, C., Birke, M., Banks, D., Filzmoser, P. and Frengstad, B. (2015) Inorganic chemical quality of European tap-water: 2. Geographical distribution. Applied Geochemistry 59, 211-224.

Fuller, R.M., Smith, G.M., Sanderson, J., Hill, R. and Thomson, A. (2013) The UK Land Cover Map 2000: construction of a parcel-based vector map from satellite images. The Cartographic Journal.
Funari, E. and Testai, E. (2008) Human health risk assessment related to cyanotoxins exposure. Critical reviews in toxicology 38(2), 97-125.
Global Water Intel. Market Profile: The water market in England and Wales to 2020. https://www.globalwaterintel.com/client_media/uploaded/Chantal/market%20profile%20sample.pdf, 2014.
Gooddy, D.C., Ascott, M.J., Lapworth, D.J., Ward, R.S., Jarvie, H.P., Bowes, M.J., Tipping, E., Dils, R. and Surridge, B.W.J. (2017) Mains water leakage: Implications for phosphorus source apportionment and policy responses in catchments. Science of The Total Environment 579, 702-708.
Gooddy, D.C., Lapworth, D.J., Ascott, M.J., Bennett, S.A., Heaton, T.H.E. and Surridge, B.W.J. (2015) Isotopic Fingerprint for Phosphorus in Drinking Water Supplies. Environmental Science & Technology 49(15), 9020-9028.
Hayes, C. (2010) Best practice guide on the control of lead in drinking water, IWA Publishing.
Hayes, C. (2012) Internal Corrosion of Water Supply Systems, IWA Publishing, London.
Hayes, C., Incledion, S. and Balch, M. (2008) Experience in Wales (UK) of the optimisation of ortho-phosphate dosing for controlling lead in drinking water. Journal of water and health 6(2), 177-185.
Haygarth, P.M., Jarvie, H.P., Powers, S.M., Sharpley, A.N., Elser, J.J., Shen, J., Peterson, H.M., Chan, N.-I., Howden, N.J. and Burt, T. (2014) Sustainable phosphorus management and the need for a long-term perspective: The legacy hypothesis. Environmental Science & Technology 48(15), 8417-8419.
Hilton, J., O'Hare, M., Bowes, M.J. and Jones, J.I. (2006) How green is my river? A new paradigm of eutrophication in rivers. Science of The Total Environment 365(1–3), 66-83.
Holman, I., Whelan, M., Howden, N., Bellamy, P., Willby, N., Rivas-Casado, M. and McConvey, P. (2008) Phosphorus in groundwater—an overlooked contributor to eutrophication? Hydrological Processes 22(26), 5121-5127.
Irish Water (2015) Draft Water Services Strategic Plan: A Plan for the Future of Water Services, Irish Water Dublin.
Jarvie, H.P., Neal, C. and Withers, P.J.A. (2006) Sewage-effluent phosphorus: A greater risk to river eutrophication than agricultural phosphorus? Science of The Total Environment 360(1), 246-253.
Johnson, A.C., Acreman, M.C., Dunbar, M.J., Feist, S.W., Giacomello, A.M., Gozlan, R.E., Hinsley, S.A., Ibbotson, A.T., Jarvie, H.P., Jones, J.I., Longshaw, M., Maberly, S.C., Marsh, T.J., Neal, C., Newman, J.R., Nunn, M.A., Pickup,

R.W., Reynard, N.S., Sullivan, C.A., Sumpter, J.P. and Williams, R.J. (2009) The British river of the future: How climate change and human activity might affect two contrasting river ecosystems in England. Science of The Total Environment 407(17), 4787-4798.

Killick, R. and Eckley, I.A. (2014) changepoint: An R Package for Changepoint Analysis. 2014 58(3), 19.

Kinniburgh, J.H. and Barnett, M. (2010) Orthophosphate concentrations in the River Thames: reductions in the past decade. Water and Environment Journal 24(2), 107-115.

Lambert, A. (1994) Accounting for Losses: The Bursts and Background Concept. Water and Environment Journal 8(2), 205-214.

Li, H., Liu, J., Li, G., Shen, J., Bergström, L. and Zhang, F. (2015) Past, present, and future use of phosphorus in Chinese agriculture and its influence on phosphorus losses. AMBIO 44(2), 274-285.

Liu, S., Gunawan, C., Barraud, N., Rice, S.A., Harry, E.J. and Amal, R. (2016) Understanding, Monitoring, and Controlling Biofilm Growth in Drinking Water Distribution Systems. Environmental Science & Technology 50(17), 8954-8976.

Marsh, T. and Hannaford, J. (2008) UK hydrometric register. A catalogue of river flow gauging stations and observation wells and boreholes in the United Kingdom together with summary hydrometric and spatial statistics, Centre for Ecology & Hydrology.

Merrett, S. (2007) The Thames catchment: a river basin at the tipping point. Water Policy 9(4), 393-404.

Met Office (2010) UK Climate Projections: Briefing report, Met Office,, Exeter, UK.

Met Office (2016) Climate Summaries.

Miettinen, I.T., Vartiainen, T. and Martikainen, P.J. (1996) Contamination of drinking water. Nature 381(6584), 654-655.

Moatar, F., Abbott, B.W., Minaudo, C., Curie, F. and Pinay, G. (2017) Elemental properties, hydrology, and biology interact to shape concentration-discharge curves for carbon, nutrients, sediment, and major ions. Water Resources Research 53(2), 1270-1287.

Mockler, E.M., Deakin, J., Archbold, M., Gill, L., Daly, D. and Bruen, M. (2017) Sources of nitrogen and phosphorus emissions to Irish rivers and coastal waters: Estimates from a nutrient load apportionment framework. Science of The Total Environment 601-602, 326-339.

Morée, A.L., Beusen, A.H.W., Bouwman, A.F. and Willems, W.J. (2013) Exploring global nitrogen and phosphorus flows in urban wastes during the twentieth century. Global biogeochemical cycles 27(3), 836-846.

Murphy, J.M., Sexton, D.M.H., Jenkins, G.J., Booth, B.B.B., Brown, C.C., Clark, R.T., Collins, M., Harris, G.R., Kendon, E.J., Betts, R.A., Brown, S.J., Humphrey, K.A., McCarthy, M.P., McDonald, R.E., Stephens, A., Wallace, C., Warren, R., Wilby, R. and Wood, R.A. (2009) UK Climate Projections Science Report: Climate Change Projections, p. 192, Meteorological Office Hadley Centre, Exeter, UK.

Ofwat. Water sector overview. http://www.ofwat.gov.uk/regulated-companies/ofwat-industry-overview/, 2017.

Olyaie, E., Banejad, H., Chau, K.-W. and Melesse, A.M. (2015) A comparison of various artificial intelligence approaches performance for estimating suspended sediment load of river systems: a case study in United States. Environmental Monitoring and Assessment 187(4), 189.

Pocock, S.J., Shaper, A.G., Ashby, D., Delves, H.T. and Clayton, B.E. (1988) The Relationship between Blood Lead, Blood Pressure, Stroke, and Heart Attacks in Middle-Aged British Men. Environmental Health Perspectives 78, 23-30.

Pohlert, T. (2018) Package 'trend'.

Powers, S.M., Bruulsema, T.W., Burt, T.P., Chan, N.I., Elser, J.J., Haygarth, P.M., Howden, N.J.K., Jarvie, H.P., Lyu, Y., Peterson, H.M., Sharpley, A.N., Shen, J., Worrall, F. and Zhang, F. (2016) Long-term accumulation and transport of anthropogenic phosphorus in three river basins. Nature Geosci 9(5), 353-356.

R Development Core Team (2016) R: A language and environment for statistical computing, R Foundation for Statistical Computing, Vienna, Austria.

Saul, A., Shepherd, W. and Boxall, J. (2007) World Environmental and Water Resources Congress 2007, American Society of Civil Engineers, Tampa, Florida, United States.

Sewage World. Water and Wastewater Treatment Plant Directory. http://www.sewage.net/, 2016.

Sharpley, A. (2016) Managing agricultural phosphorus to minimize water quality impacts. Scientia Agricola 73, 1-8.

Sharpley, A., Jarvie, H.P., Buda, A., May, L., Spears, B. and Kleinman, P. (2013) Phosphorus Legacy: Overcoming the Effects of Past Management Practices to Mitigate Future Water Quality Impairment. Journal of Environmental Quality 42(5), 1308-1326.

Southeast Water. WRMP Library. http://www.southeastwater.co.uk/about-us/our-plans/water-resources-management-plan/wrmp-library. Snodland, UK: Southeast Water, 2014.

Sutton and East Surrey Water. Final Water Resources Management Plan. https://www.waterplc.com/userfiles/file/WRMP_Final_2014_Tables.pdf. Redhill, UK: Sutton and East Surrey Water, 2014.

Taormina, R., Chau, K.-W. and Sivakumar, B. (2015) Neural network river forecasting through baseflow separation and binary-coded swarm optimization. Journal of Hydrology 529, 1788-1797.
Tchobanoglous, G. and Burton, F.L. (2013) Wastewater Engineering: Treatment and Resource Recovery, McGraw-Hill, New York, USA.
Thames Water. Water Resources Management Plan 2015 - 2040. http://www.thameswater.co.uk/about-us/5392.htm. Reading, UK: Thames Water, 2014.
UK Water Industry Research Ltd (2007) Managing Seasonal Variations in Leakage, UK Water Industry Research Ltd, London.
UK Water Industry Research Ltd (2012) Alternatives to Phosphate for Plumbosolvency Control UK Water Industry Research Ltd, London.
UK Water Industry Research Ltd (2013) Effect of Weather on Leakage and Bursts, UK Water Industry Research Ltd,, London.
Vollenweider, R.A. (1968) Scientific fundamentals of the eutrophication of lakes and flowing waters, with particular reference to nitrogen and phosphorous as factors in eutrophication.
Wang, W.-c., Xu, D.-m., Chau, K.-w. and Lei, G.-j. (2014) Assessment of River Water Quality Based on Theory of Variable Fuzzy Sets and Fuzzy Binary Comparison Method. Water Resources Management 28(12), 4183-4200.
Water Technology. Latest Industry Projects In Europe. http://www.water-technology.net/projects/region/europe/, 2016.
White, P.J. and Hammond, J.P. (2009) The sources of phosphorus in the waters of Great Britain. Journal of Environmental Quality 38(1), 13-26.
World Bank (2006) The Challenge of Reducing Non-Revenue Water (NRW) in Developing Countries, World Bank, Washington DC.
Worrall, F., Howden, N.J.K., Burt, T.P. and Jarvie, H.P. (2015) The fluvial flux of phosphorus from the UK 1974 – 2012: where has all the phosphorus gone?, Vienna, Austria.
Zhang, Q. (2018) Synthesis of nutrient and sediment export patterns in the Chesapeake Bay watershed: Complex and non-stationary concentration-discharge relationships. Science of The Total Environment 618, 1268-1283.
Zhang, Q., Harman, C.J. and Ball, W.P. (2016) An improved method for interpretation of riverine concentration-discharge relationships indicates long-term shifts in reservoir sediment trapping. Geophysical Research Letters 43(19), 10,215-210,224.