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The fate of phosphate (and chlorine) in drinking water distribution systems

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ABSTRACT

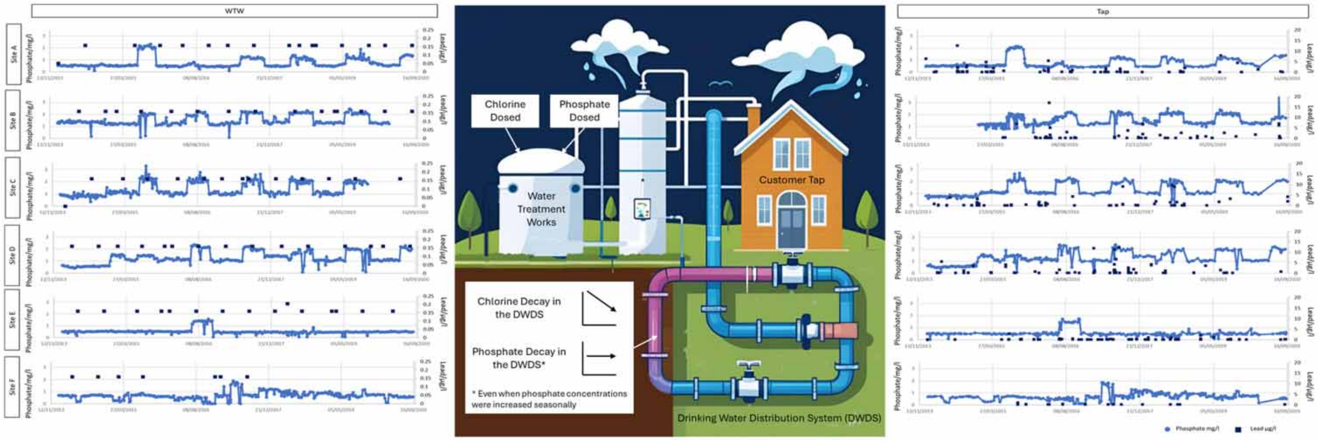
Chemicals are added to treated drinking water to protect water quality during long residence times within ageing drinking water distribution systems (DWDSs). The impacts and benefits of this, phosphate dosing to mitigate lead exposure risks in particular, in operational systems, are not fully understood, and hence dosing practices may not be optimal. Regulatory sampling data from the water treatment works (WTWs) outlet and at customers' taps from six contrasting systems for 6 years were analysed (2014–2020). Results showed the expected variation in chlorine decay throughout the DWDS as a function of the source water, network infrastructure and residence time. Phosphate concentrations had no measurable change detected from the WTW to the customer tap, irrespective of the seasonal dose. As phosphate concentrations at the WTW increased seasonally, at times doubling, the phosphate concentrations at the tap followed suit, questioning the effectiveness of this increased seasonal dosing. This is significant given the increasing costs, decreasing availability and wider environmental concerns around phosphate.

Key words: chlorine, drinking water distribution chemicals, drinking water distribution systems, lead, phosphate

HIGHLIGHTS

- Seasonal increases of phosphate were applied during summer months in the case study water company for lead control in warmer temperatures.
- Phosphate concentrations showed no measurable reduction from water treatment works to customer taps across six diverse networks.
- Chlorine decay varied substantially between networks, unlike phosphate.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Drinking water distribution systems (DWDSs) are vast, complex networks that distribute consistently high-quality drinking water from water treatment works (WTWs) to customer taps. There is over 350,000 km of DWDS pipe in England and Wales, comprising a diverse range of materials, diameters, ages and complexity. These DWDSs are the vital last stage in supplying safe, clean drinking water, essential for public health, society and the economy. To help maintain drinking water quality on this journey, distribution network chemicals are dosed. To limit microbial regrowth and protect against pipe ingress, a disinfection residual such as chlorine is maintained. To mitigate the presence of lead pipes, fittings and fixtures, an orthophosphoric acid dose is applied. While there has been a wealth of research regarding chlorine dose, concentrations and impacts, the wider role and impacts of phosphate within the vast complexity of DWDSs are not fully understood. Most research efforts have focused on lead and lead release rather than the wider impact of phosphate dose.

This research aimed to understand the role and impact of phosphate in operational DWDSs by studying the change in phosphate concentrations between WTWs and consumer taps. Particular focus was given to understanding if and what benefits resulted from changes in seasonal dosing practices, and if positive or adverse effects could be observed, such as phosphate acting as a nutrient to increase microbial numbers. Chlorine data were also studied as a comparison to the phosphate results.

1.1. Background

1.1.1. Lead and phosphate

Although it was banned to lay fresh lead pipes in 1969 in the UK, old lead pipes are still present, especially in houses built before this date. Lead was a common pipe material because of its durability, malleability and strength, which made pipe installation quick and simple (Van der Leer 1999). It is estimated that there is lead presence in ~40% of UK homes, meaning that ~17 million people are still supplied by lead pipes (European Institution for Testing, Inspection and Certification 1998). Lead can be present in both customer-owned and utility-owned pipes. One major water utility has an estimated 1.2 million lead company-owned communications pipes, which would cost ~£50 billion to replace and take 120 years (WatNet 2022). Lead can also be present within the home from solder, fittings and fixtures (World Health Organization 2023).

Prolonged contact with lead can have serious consequences for human health. One of the impacts of lead toxicity that has been highly studied is an adverse impact on the mental development of children. Blood lead concentrations as low as 5 μg/dl, once thought to be a 'safe level', can cause reduced IQ and behavioural changes in children (World Health Organization 2023). It is now agreed that there is no safe blood level of lead (Betts 2012). The responsibility for lead exposure is with everyone; water is not the only source.

Lead can be accumulated in the form of particulate lead due to lead flaking from old lead pipes or galvanic corrosion products, as well as dissolved lead from lead carbonates on the pipe wall. Without management, lead would often be present in tap water because of the corrosive effects of water on household plumbing systems, which contain lead pipes, solder or fittings. To prevent the public from ingesting lead and to adhere to strict regulations (<10 μg/l lead at the consumer tap), phosphate, in the form of orthophosphoric acid (H_3PO_4) or orthophosphate (PO_4^{3-}), is dosed at ~95% of all WTW in the UK (International Water Association 2016; Lamb 2020). These phosphate-based chemicals convert some of the lead carbonate in the corrosion film to lead phosphate, which is less soluble (International Water Association 2010a, b). In this way, an insoluble surface of lead phosphate is formed, creating a protective layer around lead pipes to prevent lead leaching (European Commission 2016).

The formation process of the protective lead phosphate layer consists of a complex set of simultaneous reactions and equilibrium constant expressions, driven by concentration gradients of lead phosphate, lead carbonate and the relative concentrations of orthophosphate, carbon and bicarbonate ions (Wenzel 2005; International Water Association 2010a, b). A commonly used practice in the UK water utility companies is to apply an orthophosphate dose of 1.0 mg/l for 1 year, followed by a continuous lower dose (0.5–2.0 mg/l) to indefinitely maintain the layer area (Hayes *et al.* 2014). A continual supply of phosphate is required to maintain the plumbosolvency effect. If the dose is lowered, a new equilibrium state is established, meaning that the pipe wall precipitates disperse and the phosphate gradually re-enters the bulk water (Brandt *et al.* 2017). The time it takes for the plumbosolvency effects to cease following dose reduction or discontinuation is uncertain, ranging from a few months to several years (Hayes *et al.* 2008).

Plumbosolvency is governed by a combination of thermodynamics and kinetic processes, meaning that a number of factors can affect the formation of the protective layer, including pH, alkalinity, orthophosphate and water temperature (Brandt *et al.*

2017). The International Water Association (IWA) (2010a, b) reported that in some studies, lead concentrations halved when temperatures decreased from 25 to 12 °C. Due to results such as these, some water companies increase phosphate doses in the summer and reduce doses in the winter to account for lead solubility increasing with increasing water temperatures (Masters Welter & Edwards 2016). Multiple reports designed for operators confirm this practice, including the IWA (2010a, b) Guide for Small Community Water Suppliers and Local Health Officials on Lead in Drinking Water, UK Water Industry Research (UKWIR) (2012) The Alternatives to Phosphate for Plumbosolvency Report and The IWA (2010a, b) Best Practice Guide on the Control of Lead in Drinking Water.

The impetus of having just sufficient concentrations of phosphate to have a favourable equilibrium for lead control is compounded by the ambition to preserve phosphate. Phosphate is a non-renewable resource that is mined from phosphate rock reserves. There are an estimated 67,700 million tonnes of global phosphate rock remaining, which may be depleted as soon as in the next 50–100 years (Cordell *et al.* 2009). As such, phosphate concentrations need to be carefully understood and considered.

The application of phosphate may also have unintended impacts on the DWDS. For example, phosphate has been found to effectively reduce the solubility of metal piping beyond lead, such as copper pipes and iron pipes (Rosales *et al.* 2020). A further example is that little is known about the impact of the phosphate dose on the ecology of microorganisms that persist in DWDS. Most microorganisms utilise phosphate for growth, with even low concentrations of 1 μg/l PO_4^{3-} able to increase and support intense levels of biological activity (Miettinen *et al.* 1997). Douterelo *et al.* (2020) noted differences in the taxonomic composition of both bacteria and fungi when a phosphate dose was and was not applied.

It is not known what the impact of the application of a seasonal phosphate dose has on the DWDS, for instance, the microbial impact of this practice. While several water industry guidance documents could be seen to be recommending this course of action to help prevent toxic lead leaving consumer taps, there is little research into the effectiveness of this seasonal phosphate dosing, which is of concern because increasing the phosphate dose is expensive and may not be sustainable due to diminishing phosphate reserves. Water companies have done many internal reviews of lead dose and the impacts of phosphate on a site-by-site basis, but these data are rarely published in the public domain. Concentrations of phosphate at the consumer tap are infrequently published, as the main focus is on lead presence at the tap.

1.1.2. Chlorine

A secondary disinfectant is frequently applied at the end of the WTW, in addition to the primary disinfection completed earlier on in the treatment train, within the WTW itself. This disinfectant residual is commonly maintained in distributed water to limit microbial regrowth and mitigate against rare microbial contamination events such as cross connection, back siphoning and pipe ingress within the DWDS. Often applied is a free chlorine (HOCl) dose of 0.5–1.0 mg/l, or in some systems, combined chlorine (chloramination, a combination of HOCl and ammonia) is used. While a chlorine residual is not a requirement of WHO, EU or UK regulation, it is a recommendation and water utilities aim to maintain a measurable residual at the customer tap (World Health Organization 1984; Drinking Water Inspectorate 2016; European Commission 2016).

Chloramine use is increasing in the UK water industry mainly because of concerns regarding the formation of disinfection by-products (DBPs), some of which can be carcinogenic. When compared with chlorine use, chloramines have been found to reduce the formation of one type of DBP, trihalomethanes (THMs) by 40–60% (Kirmeyer 2004). Chloramine can also be favoured for its increased persistence and minimised taste and odour concerns, including those that are chlorinous (Westbrook & Digiano 2009). Although it is not as effective at disinfecting water as free chlorine, the majority of studies agree that the persistence of chloramine makes it better suited than chlorine for the disinfection of biofilms (Kirmeyer 2004).

While chloramine has been found to have a greater persistence than chlorine, both chloramine decay and chlorine decay occur within the DWDS. The longer water is in contact with the DWDS material, and so the higher the age of the water, the higher the propensity for disinfection residual to be decayed and water quality to be impacted (Rossman Clark & Grayman 1994; Machell & Boxall 2014).

While it is important to have a good understanding of water age in DWDS, it is not the sole indicator for disinfectant decay. This process is dependent on multiple factors including: disinfection method used, maintenance of the residual in the DWDS, variety and concentration of nutrients present in the water, source water type, treatment processes used, DWDS length, pipe material and the temperature of the water (Momba *et al.* 2000). As different DWDS have different characteristics, different DWDS consume chlorine at different rates, and it is important to conduct research into a variety of case study areas.

2. METHODS

Central to addressing the aims of this research was to collect sufficient data from operational water supply systems to enable meaningful conclusions to be drawn. In particular, data to explore the changes in concentrations of chemicals during the transit of water through DWDSs. To best achieve this, data from a selection of DWDSs with different contrasting conditions, from a multi-year period, were obtained. This provided greater insight, including attribution of cause and effects, than collecting data from across a random larger selection of DWDSs. The key data required was:

- Phosphate at the WTW and consumer taps to observe dosing levels and utilisation within the DWDS.
- Lead at the WTW and consumer taps to confirm the presence of lead pipes, fittings and fixtures.
- Chlorine at the WTW and taps to assess the DWDS variety through varying levels of chlorine decay, as well as to provide a better understanding of comparison to the phosphate results.
- Bacterial count data to observe changes occurring in the DWDS and the possible impacts of phosphate and chlorine on this.

2.1. Sample data

The required data were available from laboratory analysis of discrete samples, in accordance with regulatory requirements. Sample analysis was completed following the Standing Committee of Analysts Blue Book methods, with analysis occurring at a UKAS-accredited laboratory to the Drinking Water Testing Specification. Sample replication is not required in these regulations.

Sampling at the final water of a WTW is generally daily or weekly, depending on the parameter and size of the WTW, and is obtained from a designated regulatory compliance sample tap. Hence, well-structured data clearly defining water quality entering DWDSs are available. The same is not true of the consumer tap data, as they were procured from random daytime sampling of different properties at different times, as per regulatory requirements. Due to the lack of spatial and temporal consistency of the consumer tap data, it is a complex dataset not suitable for statistical analysis, including significance. Despite this, a substantive dataset was collected, with reliable time stamping, enabling the use of subtle visual correlations that are time-based. Importantly, each customer tap sample was analysed for both phosphate, lead and chlorine; hence these are directly comparable for each tap sample.

2.2. DWDS selection

Six public water supply zones or DWDSs were carefully selected to provide contrasting conditions, each spanning a period of 6 years. DWDSs were selected to ensure that there was sufficient historic data availability, as operational DWDS that were new, changed or are not currently operational would have poor or inconsistent data availability for comparison with other DWDSs and for comparison over time. Further key criteria were different source water types, different secondary disinfection methods and a combination of phosphate dosing practices. Due to the nature of this variation, the selected DWDSs unavoidably also had a range of other characteristics, including residence times and size. Details of these are captured in Table 1.

Table 2 provides further details of the phosphate dosing achieved in each DWDS. The table also includes the estimation of ‘lead risk’ assigned to each by the associated water company, based on historic sample data. From this, it can be seen that Sites A to D had seasonal dosing planned and implemented based on medium lead risk or high lead risk. The implementation of seasonal summer increased seasonal dosing was not consistently applied every year, enabling some exploration of the effects of this. Table 2 also shows that Sites E and F did not have a seasonal phosphate dose. Where a seasonal dose was applied, the temperature of tap water was used to begin the summer dosing period (for instance, a rise above 15 °C in late May–early June and then a decrease in October).

3. RESULTS

3.1. Chlorine decay in the DWDS

Figure 1 shows the chlorine concentration data for each DWDS for the 6-year period. There was an expected spread in the data at the WTW due to changes over the 6 years.

Both free and total chlorine are presented in Figure 1, with the use of chloramine rather than free chlorine residual at Sites A and C being evident from the much lower free chlorine presence at both the WTW and the tap.

Table 1 | Details of the six case study DWDS selected

Site	PWSZ population fed	Source water type	Water treatment process	Network chemicals	Water age (days)	PWSZ network length (m)	Predominant network material	Average total chlorine (mg/l)	Average 3 day colony count (no/ml)	Average 2 day colony count (no/ml)	Average total lead (μg/l)
A WTW	38,082	SW	GAC, slow sand filters	NH_2Cl, P	≥ 6	285,000	52% plastic 41% iron 6% cement	1.008	0.723	0.841	0.153
A Tap								0.541	2.302	7.211	0.651
B WTW	18,087	SW	Ozone, RGF, GAC	HOCl, P	4	860,000	53% concrete 30% plastic 16% iron	1.014	0.272	0.114	0.160
B Tap								0.406	0.791	2.847	0.923
C WTW	16,539	SW Blend	Ozone, RGF, GAC	NH_2Cl, P	≥ 6	280,000	60% plastic 24% iron	1.114	9.784	0.785	0.160
C Tap								0.436	2.722	6.591	0.668
D WTW	18,390	SW/GW Blend	Ozone, GAC	HOCl, P	≥ 6	716,000	40% plastic 27% iron 15% steel	1.020	0.145	0.405	0.160
D Tap								0.652	1.949	38.410	0.881
E WTW	19,685	GW	RGF	HOCl, P	3	158,000	49% plastic 36% cement 14% iron	0.744	0.160	0.579	0.163
E Tap								0.469	0.370	0.691	0.242
F WTW	8,592	GW	NA	HOCl, P	2	115,000	56% plastic 25% iron 19% cement	0.509	0.049	0.054	0.160
F Tap								0.439	0.274	2.883	0.214

SW, surface water; GW, groundwater; GAC, granular activated carbon; RGF, rapid gravity filters; PWSZ, public water supply zone; NH_2Cl, chloramine; P, phosphate; HOCl, free chlorine; NA, not applicable.

Table 2 | Details of the seasonal phosphate dosing implemented at each of the six selected DWDS and the lead risk categorisation that was used to inform this dose

	Average achieved phosphate dose (mg/l)			
DWDS	Non-summer	Summer	Lead risk	Lead risk meaning
A	0.599	1.10	Medium	Any PWSZ with >2 and <5% samples >5 μg/l
B	1.35	1.79	Medium	Any PWSZ with >2 and <5% samples >5 μg/l
C	1.13	1.8	High	Any PWSZ with >5% samples >5 μg/l
D	1.13	1.57	Medium	Any PWSZ with >2 and <5% samples >5 μg/l
E	0.475	0.60	Very Low	No samples with >5 μg/l lead
F	0.742	NA	Very Low	No samples with >5 μg/l lead

NA, not applicable.

Chlorine concentrations were reduced from the WTW to the customer taps in each case study area, clearly showing expected chlorine decay in each DWDSs. The amount of decay and spread in chlorine concentration data in the customer tap data was greater than at the WTW, as expected, due to the random selection of customer tap sampling points.

A comparison of the observed chlorine decay between the different DWDSs showed a complex picture:

- Sites E and F had the least decay. These areas were groundwater-sourced, had the lowest residence times and the smallest DWDS.
- In contrast, Site B had nearly as low an average residence time as Sites E and F but exhibited among the greatest chlorine decay. This area was surface water-fed and with a larger DWDSs.

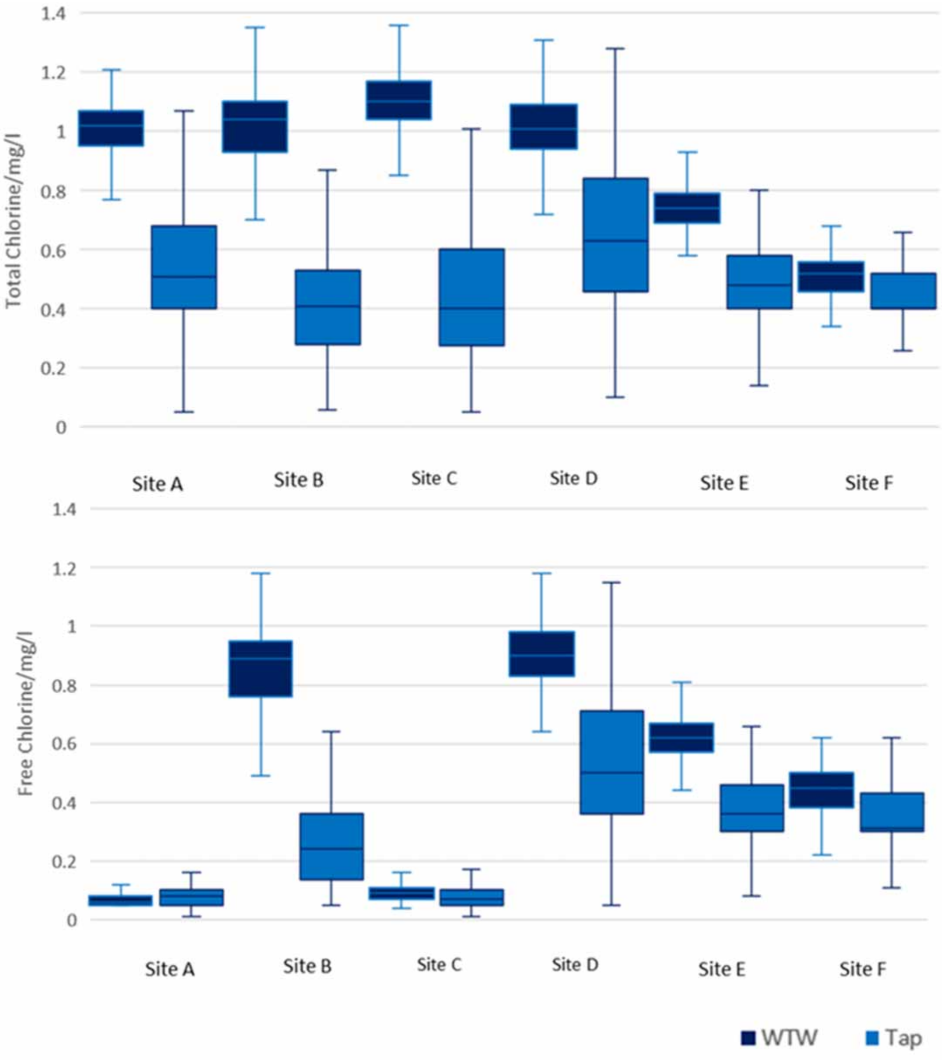


Figure 1 | Free and total chlorine concentrations at the WTW final water and customer taps. Box and whisker plots show the median, 25th and 75th percentiles for the 6 years of data at each DWDS. Sample numbers for Sites A–F: Total chlorine WTW 1307, 2487, 2561, 2518, 633, 189. Total chlorine tap 1185, 963, 825, 889,777,415. Free chlorine WTW 1307, 2487, 2561, 2518, 635, 189. Free chlorine tap 1183, 962, 825, 889, 777, 415.

- Sites A and C exhibited significant chlorine decay, with Site C the greatest of all six. They were a little larger than Sites E and F, but with greater residence times.
- Sites B and D were the largest DWDSs but had very different average residence times. Site D had a greater residence time but has noticeably less chlorine decay than Site B.
- Sites A and C had a disinfection method of chloramine rather than free chlorine. Despite this, Site C showed the greatest decay of any DWDS, without being the DWDS with the largest or greatest residence time. This showed no observable persistence of the disinfection residual from the use of chloramines, in contrast to previously reported data.

These results confirm that chlorine decay is a complex interaction of source water, DWDS infrastructure and residence time, as well as other factors, such as temperature.

The range of chlorine concentrations observed at the taps at each DWDS showed that the random tap sampling was covering a range of different DWDS residence times and conditions. Chlorine decay was evident despite this random sampling throughout the length and breadth of these diverse DWDSs. This provides evidence that the expected complex reactions and interactions were occurring within these DWDSs and being captured and represented in the available data.

It could be expected from these data that a similar trend should be seen in phosphate usage and concentrations throughout the DWDS.

3.2. Phosphate decay in the DWDS

Figure 2 presents the WTW and tap phosphate concentration data, in the same form as the chlorine data were presented in Figure 1. Little or no change was observed in phosphate concentrations between WTW and customer taps. Overall averages showed a very slight increase to 1.07 mg/l at the taps compared with the dose applied at the WTW 0.988 mg/l, but this was not significant.

There was no consistent or significant change in the average and range of values of phosphate concentrations between the WTW and taps for each DWDS. Only Site F showed a possible change, a slight increase from WTW to taps. In stark contrast to the chlorine data, there was no measurable reduction in phosphate concentrations from WTW to tap. This was irrespective of the DWDS conditions presented in Table 1, showing that the factors of source water type, DWDS infrastructure and size and residence time did not impact phosphate utilisation.

3.3. The impact of seasonal phosphate dose on tap phosphate concentrations

Figure 3 explored the phosphate data further, presenting it as a time series to explore any impacts of the seasonal dosing described in Table 2. The periods of seasonal dosing can be clearly seen in Figure 3. While seasonal dosing was delivered for Sites A – D, with variations between years, it can also be observed that there were periods of increased dosing at Sites E and F, despite this not being the standard procedure for these DWDSs.

Clearly demonstrated by Figure 3 was that there was no measurable change in phosphate concentrations between the consumer tap and the WTW. The time series data also revealed that there was no decay or release as a function of the seasonal dosing changes, with the changes in seasonal phosphate dose at the WTW (left) exactly mirrored in the tap data (right). Plotting the data on the same axis leads to one or the other dataset being entirely obscured; hence, this is not included as a figure.

3.4. The microbial impacts of chlorine and phosphate

There is a wealth of research that states that to a certain extent, temperature increases the rates of reaction times, including chlorine decay, lead mobilisation and microbial growth. As such, Figure 4 was aimed to explore temperature change, chlorine concentration and microbial abundance over time.

Figure 4 denotes that microorganisms occurred within all DWDSs. There were occasional slight increases in the numbers of microorganisms observed in both WTW and tap data in all DWDSs, despite the presence of a disinfection residual.

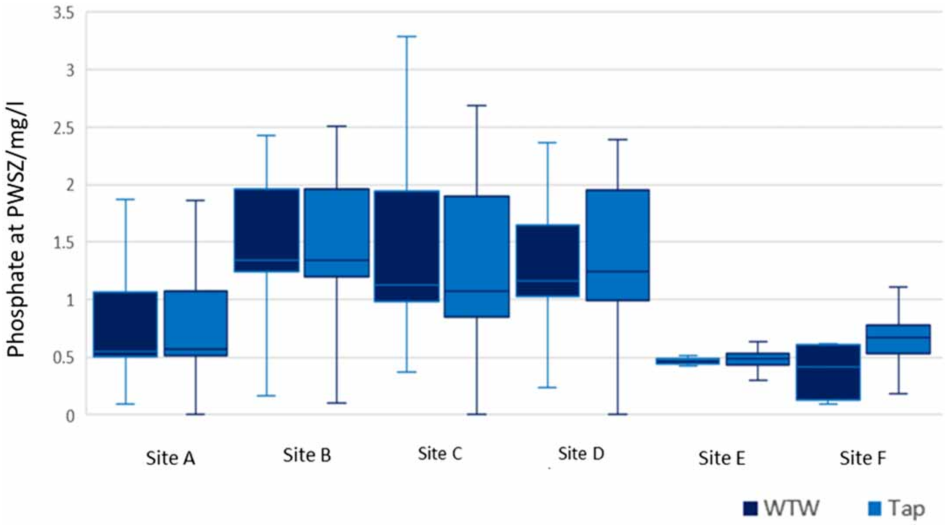


Figure 2 | Phosphate concentration data at the WTW final water and customer taps. Box and whisker plots show the median, 25th and 75th percentiles for the 6 years of data at each DWDS. Sample numbers for Sites A–F: Phosphate WTW 353, 350, 352, 347, 349, 288. Phosphate tap 314, 433, 334, 336, 288, 291.

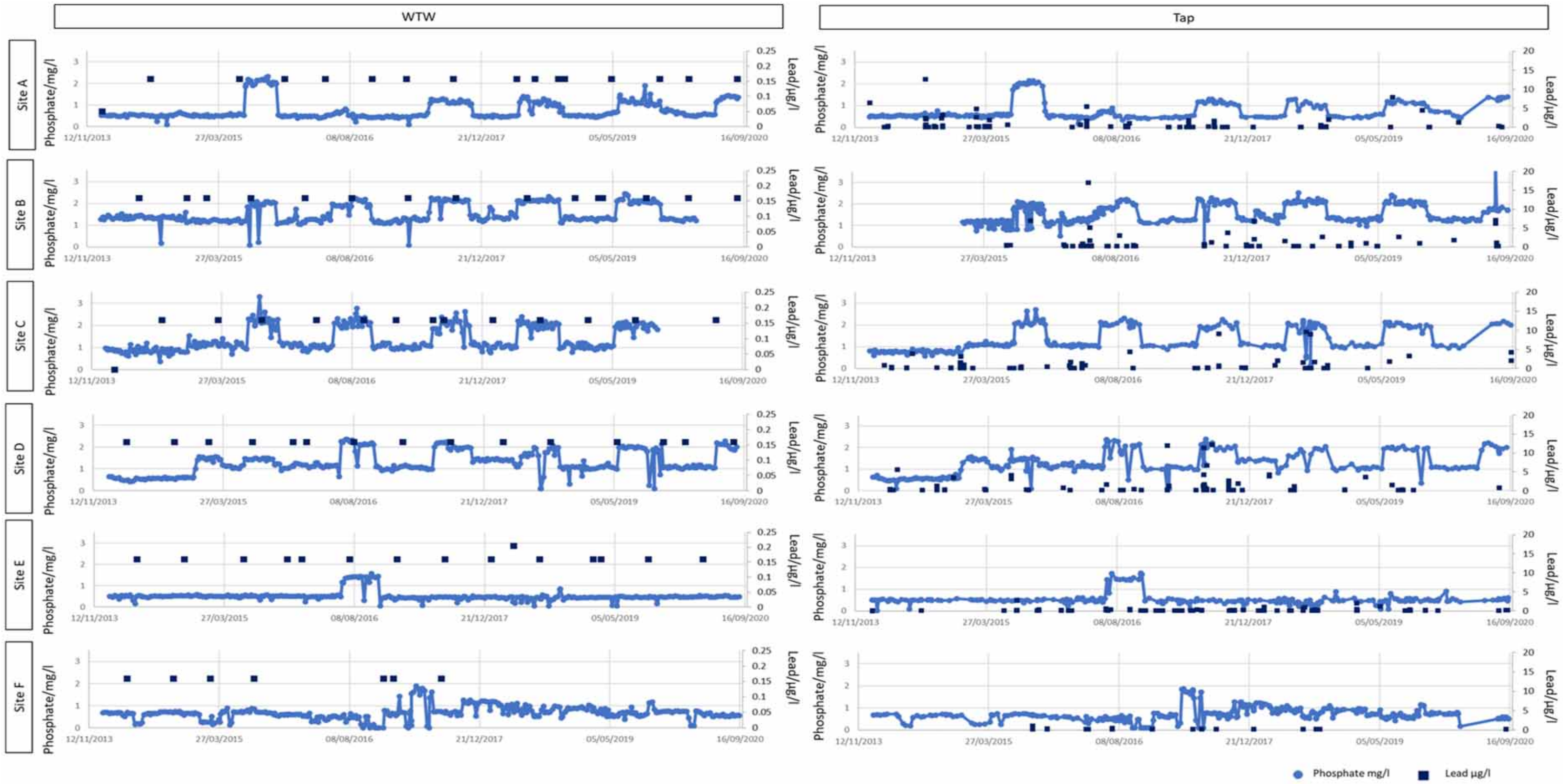


Figure 3 | Phosphate and lead data as a time series. WTW data are shown in the left column, tap data in the right. Each row of the table is a different case study site. Secondary *x*-axis for lead is 0–0.25 μg/l for the WTW data and 0–20 μg/l for the tap data; the phosphate axis is the same. Sample numbers for Sites A–F: Phosphate WTW 353, 350, 352, 347, 349, 288. Phosphate tap 314, 433, 334, 336, 288, 291. Lead WTW 16, 15, 14, 15, 16, 7. Lead tap 157, 98, 134, 139, 134, 44.

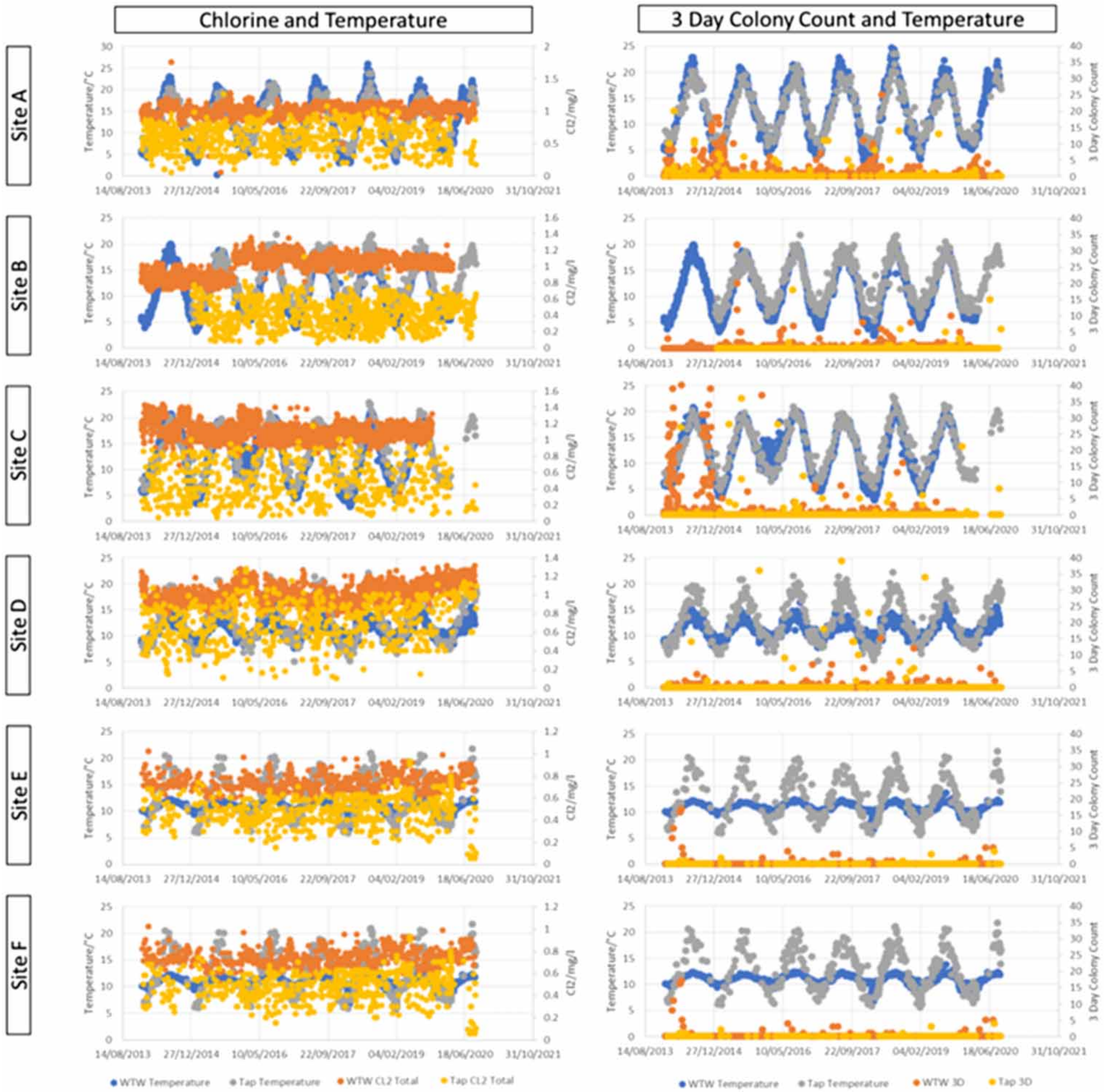


Figure 4 | Water temperature and total chlorine residual at the WTW and tap, left column. Water temperature and 3 day colony counts at the WTW and tap, left column. Each row is a DWDS. Sample numbers for Sites A–F: Total chlorine WTW 1307, 2487, 2561, 2518, 633, 189. Total chlorine tap 1185, 963, 825, 889,777,415. Temperature WTW 1207, 2461, 2460, 2466, 597, 178. Temperature tap 366, 366, 335, 408, 372, 182. 3 day colony count WTW 1256, 2466, 2503, 2320, 614, 185. 3 day colony count tap 235, 211, 241, 256, 146, 113.

No repeated seasonal pattern in the microbial numbers was observed, particularly in the DWDS. This was despite the seasonal temperature changes and despite the seasonal increases in phosphate dose (Figure 3). This showed that within the resolution of this dataset, there was no seasonal impact of temperature or phosphate dose on microbial numbers. This finding suggests that phosphate was not the limiting nutrient in any of these DWDS.

Figure 4 also provided a more detailed view of the chlorine data. There was no obvious difference in the reduction in the residual between the free chlorine and chloramine DWDSs. A further observation is that no clear seasonal pattern was present. This could be because random customer tap sampling may have been capturing the greater effects of different infrastructure pathways and residence times within the DWDS, rather than any impacts of increased chlorine decay due to higher temperatures.

4. DISCUSSION

4.1. An exploration of chlorine dosage practices

As expected, disinfection residual decay was clearly noted between the WTW and the tap (Figure 1). The volume of data compiled and presented here is large. During the study period, 40,227 WTW samples and 16,038 tap samples, a total of 56,265 were analysed. Of these samples, there were 14,749 total chlorine samples and 14,748 free chlorine samples. However, the intensity of the tap samples was low spatially and temporally with respect to the size, complexity and continual operation of DWDSs. The clearly overserved trends in chlorine showed that the sampling coverage was sufficient to observe DWDS-wide reactions and interactions.

Due to the variety of different chlorine dosages and practices (including the use of chlorination and chloramination) and the vast array of DWDS differences between the different case study areas, as shown in Figure 1, a variety of chlorine doses were applied at the WTW. This was noted in Table 1 as total chlorine ranging from 1.114 mg/l at Site C to 0.509 mg/l at Site F. Despite these differences in chlorine concentrations applied at the WTW, total chlorine concentrations at the tap were found to be relatively similar to each other, regardless of the case study site (0.406–0.652 mg/l) (Table 1).

It could be said that chlorine was well understood in the DWDS because more chlorine samples were taken than phosphate and lead samples, in accordance with regulatory requirements. For example, the tap of Site A had 1,185 total chlorine and 1,183 free chlorine samples taken during the study period, but only 317 phosphate samples and 157 lead samples.

It was interesting to observe that the management of four of these six DWDS included seasonal change of phosphate dose due to expected temperature-related change in plumbosolvency, but that these same systems did not have a similar management strategy for seasonal increase in chlorine dose. As stated, chlorine decay is known to be influenced by water temperature, as is the microbial activity of the water to which chlorine is dosed to minimise (Momba *et al.* 2000).

4.2. An exploration of phosphate dosage practices

Figure 3 indicates that there was no lag between a change in phosphate concentration at the WTW and the phosphate concentration detected at the tap. A lag might be expected in the presence of lead pipes because the protective surface layers that form are a function of the phosphate concentration gradient from the bulk water, as described in the literature review. Numerous potential reasons as to why phosphate concentrations did not appear to change between the WTW and the tap have been discussed below.

4.2.1. There was no lead within the DWDS studied to interact with the dosed phosphate

Table 1 did not show lead to be a predominant pipe material in any of the case study DWDS, as expected, as these data are from the company asset database, not including supply or communication pipes. However, Figure 3 detailed that all case study areas had an increase in lead concentrations from the WTW to the tap, indicating that lead was present in all of the studied DWDSs. It must be noted, however, that lead samples at the WTW were highly limited, with Site E having only seven regulatory samples during the 6-year case study period. Regardless, it is interesting that lead presence was detected across all DWDSs because of the way that lead is sampled. For instance, for a population of 90,000 people, only 8 regulatory lead samples are required to be taken per year, which equates to 0.00889% of the population being tested. As the lead concentration in tap water is spatially and temporally dispersed, it is highly unlikely that lead presence will be detected with these sampling frequencies in this one discrete sample snapshot of all the water customers consume (Drinking Water Inspectorate 2016). For this reason, most water companies undertake enhanced sampling to increase the number of random daytime samples. In addition to this, samples are also taken at the request of customers in response to concerns. However, such data are necessarily biased and non-uniform, so it was not included in this analysis.

4.2.2. The phosphate dose changed too quickly

It could be that the time period between the summer dosing period and the non-summer dosing period was too short to form a new equilibrium state. As stated, the time it takes for the plumbosolvency effects to cease following dose reduction or discontinuation is uncertain, ranging from a few months to several years (Hayes *et al.* 2008).

4.2.3. Any change in phosphate concentration may have been less than the measurement resolution

It is possible that no measurable lag between phosphate concentrations at the WTW and the tap was detected, as phosphate levels may have been below the instrument's detection limit and therefore not captured by the analytical method.

4.2.4. Too high a seasonal dose of phosphate was used

As previously noted, operational reports are in circulation which confirm the practice of seasonal phosphate dosing to account for increased temperatures accelerating lead release from lead pipes, fittings and fixtures. The IWA (2010a, b) Best Practice Guide on the Control of Lead in Drinking Water refers to 'slightly higher summer dose' and 'slightly lower winter dose' but does not define the meaning of a slight dose change. Similarly, the UKWIR (2012) Alternatives to Phosphate for Plumbosolvency Report states the variation in the average dose concentration was 'within a relatively narrow range of values' but again the values themselves are not defined. Table 2 shows how, at times, some sites are doubling in phosphate dose seasonally. For example, Site A transitioning from 0.5 to 1 mg/l or Sites C and D from 1 to 2 mg/l. Further, potentially the year-round phosphate concentrations were already optimal in the selected study areas, meaning that the higher summer increase was not causing any lead impact above and beyond the already effective non-summer dose.

4.2.5. Seasonal effects, including temperature, had no impact on phosphate utilisation

This is perhaps surprising as temperature is known to affect most reactions and interactions, including the formation of the protective surfaces in lead pipes. A further explanation could be that summer ambient temperatures may be higher, but due to premise central heating during non-summer periods, there may not have been a substantial premise temperature change effect. Although Figure 4 details the water temperature of the WTW and the tap and it was found to vary seasonally. The seasonal fluctuation in temperature recorded by suppliers is a parameter that is obtained following a minimum of a 2-min tap flush. Water temperature is taken by water companies at the customer tap so technicians can ensure that they are sampling water from the mains pipe and not water that has been sitting in customer taps. Following a temperature check, other routine samples are then taken (the only exception to this is spot samples, i.e. resamples following a previous failure in that parameter). As such, the water temperature of customer taps may have been different from the water temperature recorded by samplers. As previously noted in this paper, the water temperature at the tap initiates the seasonal dosing.

The phosphate at times doubled in concentration at the WTW, but there was no measurable delay or evidence of consumption through to the tap, suggesting no utilisation of phosphate to form different concentration protective scales. This is of concern due to phosphate being an expensive and non-renewable resource. However, this finding should not be confused with or taken to infer that phosphate dosing should be stopped until and unless a comprehensive programme has been implemented and there is assurance that lead has been removed from all DWDSs, despite ownership and asset type. This is essential to help protect health. However, this work does question if there is more that could be done to reduce phosphate dosing concentrations and protect a rare non-renewable resource. The summer dose has been optimised across all WTW, including these study areas, since this data analysis period.

4.3. The impacts of distribution chemicals on microbial abundance

One area for investigation was any microbial impacts on the seasonal dosing of phosphate (Figure 3) and the continuous stable dose of chlorine (Figure 4). This was of interest because chlorine is dosed as a disinfectant to limit microbial regrowth within DWDSs, while phosphate is a nutrient source for microorganisms. It was noted that all of the case study sites had microorganisms present, despite the application of chlorine to prevent microbial growth and despite some sites having less of a phosphate dose than others. Another finding was that there was no seasonal change in microbial abundance using 3 day counts (Figure 4). Potential reasons as to why there was not a seasonal microbial boom when phosphate was dosed seasonally have been explored below.

4.3.1. 3 Day colony counts are not a sufficient measurement of bulk water microbial abundance

Much research has maintained that microbial abundance is several orders of magnitude higher when measured with absolute counting methods, such as flow cytometry, compared with culture-based quantification methods (Hassard & Whitton 2019). As Liu *et al.* (2013) said, less than 1% of the microorganisms in drinking water can be detected using colony counting. This is because only a small proportion of metabolically active microorganisms in a water sample can be grown and so detected, depending on the environmental conditions (Lautenschlager *et al.* 2013). It would be interesting for future experiments to apply different microbial abundance quantifiers. UK water companies are now using innovative techniques such as flow cytometry to improve the availability of data such as this; however, this is presently rarely focused on the customer tap.

4.3.2. Biofilm was not accounted for

It is well known that the current practice of the use of a disinfection residual in the UK is based upon its action against planktonic cells, as only bulk water is regulated because, in an operational environment, it would be very difficult to sample the pipe wall and the attached biofilm (Fish & Boxall 2018). Thus, although chlorination has been well established to be an effective disinfectant for bulk water, its impact on the biofilm is comparatively unknown, as is the impact of phosphate.

4.3.3. There were always sufficient concentrations of phosphate present for microbial growth

Within the present study, there was an average of 1.07 mg/l phosphate at the tap. Miettinen *et al.* (1997) reported that even low concentrations of 1 μg/l of phosphate are sufficient to be able to support and increase levels of microbial activity.

One interesting future piece of work would be to determine if the seasonal phosphate dose was causing a microbial community shift, either in the bulk water or in biofilm attached to the pipe surface. As Del Olmo *et al.* (2020) found, microbial community structure changes due to treatment with orthophosphoric acid can promote the presence of microorganisms that carry genes associated with the solubilisation and transport of PO_4^{3-}, including *Sphingomonas*, *Bradyrhizobium* and *Acidovorax* spp. The work of Douterelo *et al.* (2020) compared drinking water samples in a live UK network with (~1.2 mg/l) and without (~0.03 mg/l) the addition of a phosphate dose. It was found that with a phosphate dose, the relative abundance, diversity and richness were higher in fungi but lower in bacteria, compared to without a dose.

A study involving speciation would also be of interest for studying chlorine. The application of a disinfection residual may apply selective pressure that favours certain organisms over others, changing community structure in the DWDS, rather than affecting abundance as is desired from its application. For example, a study by Fish & Boxall (2018) found that the abundance of some bacterial classes varied by chlorine strength. As such, there may be more substantial differences in microbial abundance between the different case study sites and due to the application of different distribution chemicals than initially detected.

4.4. Water company practice

While this study found no direct evidence of the benefits of seasonal dosing for the six case study areas, seasonal dosing is a practice that has continued since the study period and is used presently, due to the overall company-wide improvement in lead compliance since the study period. At all water company sites (i.e. not just the six case study areas and including all investigative sampling), the average monthly failure rate for summer seasons (April–September) has decreased by 35% (±15%) between the 2007–2015 and 2015–2024 periods. The case study water companies are continuing this conservative approach of seasonal phosphate dosing, with continual review and optimisation, to see if the phosphate dose can be reduced without an impact on water quality.

5. CONCLUSIONS

This study found no measurable change in phosphate concentrations between WTW and customer taps, for 6 years of data across six different DWDSs (source water, residual type, DWDS infrastructure and residence time). Yet, the same data showed a range of decay of chlorine residual, confirming that the random sampling was capturing a range of different DWDS residence times and conditions. One explanation for this could be that the change in phosphate concentration was too subtle for the phosphate analytics used. This minimal phosphate variation is surprising as phosphate is added to form a protective surface within any lead pipes and fittings present between WTW and taps, and as such would be expected to be utilised within the DWDS to form this layer. It is known that the phosphate-enhanced surface layer within lead pipes forms and then establishes an equilibrium state. But the extent of the considerable phosphate residual at the customer taps, compared with the dose applied at the WTW, is potentially suggestive of extremely low or no utilisation or reaction with the phosphate and perhaps indicative of overdosing of phosphate, a non-renewable resource. This large phosphate tap residual occurred even though lead concentration at the customer's tap was occasionally elevated, confirming that lead was present at some locations.

Despite seasonal variation in dose, at times doubling phosphate concentrations at the WTWs of case study DWDSs, there was a universally unchanged phosphate residual at the customer tap regardless of the dosing regimen applied. Seasonal dosing was applied due to the increase in water temperature at the taps in summer months, expected to increase plumbosolvency, but no increased utilisation of phosphate was observed, which might be expected to correspond to this. No lag effect following the change in seasonal dose at the WTW was observed at the taps; this might be expected as a new equilibrium state of the layers was established. There was no change in the frequency of elevated lead concentrations either seasonally, before or after the application of a seasonal phosphate dose or in those DWDSs that did or did not receive a seasonal dose application. This suggests that there is little or no additional benefit of the seasonal dosing of phosphate at the levels being applied in these six DWDS.

Data have shown no adverse effect of the phosphate dose as a nutrient for microbial (re)growth. The minimal change in phosphate concentrations from the WTW to the tap suggests that phosphate is not having an adverse effect as a nutrient for regrowth. This suggests that phosphate is not the limiting nutrient for bacterial growth in these systems, and/or that regrowth is being limited by the disinfection residual. Although the wider impact of large and/or changing phosphate doses both within the DWDS and large phosphate residuals at the customer tap remains widely unknown.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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