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RESEARCH ARTICLE

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Watermains Leakage and Outdoor Water Use Are Responsible for Significant Phosphorus Fluxes to the Environment Across the United States

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Abstract Human activity has led to excess phosphorus (P) concentrations and the continued eutrophication of coastal and freshwaters across the United States (US). Developing more effective P management policy requires a comprehensive understanding of P sources in the environment. Public water systems across the United States widely dose water with phosphate (PO_4) in order to control the corrosion of lead and copper within their distribution networks. Using public water system PO_4 dosing facility data and target PO_4-P dosing concentrations, we estimate that PO_4 dosing added 4–14.9 kt PO_4-P yr^{-1} into the US water distribution network in 2015. Using estimates of public water supply inputs and domestic water deliveries, we estimate that 0.7–2.6, and 0.8–3.1 kt PO_4-P yr^{-1} were then lost from the network due to watermains leakage and outdoor water use, respectively. After accounting for these fluxes, we estimate that 9.3 kt PO_4-P yr^{-1} was then returned to wastewater treatment plants (WWTPs) and accounted for up to 2.7% of the national WWTP influent P load. As sources of P to the environment, lower and upper estimates of combined watermains leakage and outdoor water use PO_4-P fluxes exceeded P loads to surface waterbodies from documented point sources across 461–541 counties. The exceedance of these fluxes above other major components of the US P-budget emphasizes the need to include them in P source apportionment studies, both across the US and in other countries where public water supplies are dosed with PO_4.

Key Points:

- Leakage and outdoor water use release up to 17% and 21% of the 14.9 kt PO_4-P yr^{-1} added to US public water supply for corrosion control
- Combined leakage and outdoor water use PO_4-P fluxes exceed point source P inputs to freshwaters across 541 counties
- These fluxes should be considered in P source apportionment studies and could inform localized P management practices

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Human society is reliant on phosphorus (P) for global food production and security (Cordell & White, 2014). However, rising P demands and the nonrenewable nature of P reserves have led to global scarcity concerns (Cordell & White, 2014; Nedelciu et al., 2020; Van Vuuren et al., 2010; Yuan et al., 2018). Further, anthropogenic activity is now thought to have caused the global biogeochemical cycling of P to exceed safe planetary boundaries (Steffen et al., 2015). Excess P inputs from rural and urban environments have substantially increased P availability within fresh and coastal waterbodies around the world (Howarth, 2008; Jarvie et al., 2015; G. Metson et al., 2020; Suh & Yee, 2011). These inputs have had widespread effects on both environmental and human health (Carvalho et al., 2013; Davis & Shaw, 2006; Diaz et al., 2004), as well as significant economic costs (Garcia-Hernandez et al., 2022; Pretty et al., 2003; Sanseverino et al., 2016). This simultaneous occurrence of both P scarcity and excess, the so-called paradox of P, has made the sustainable use of P a significant global challenge (Jarvie et al., 2015; Leinweber et al., 2018).

Phosphorus pollution is a leading cause of degraded freshwater quality across the United States (US) (USEPA, 2015). High P concentrations have caused 58% of the total miles of US rivers to be rated at poor status (USEPA, 2020b), and the resulting eutrophication of the country's fresh and marine waterbodies has persisted for decades (Bricker et al., 2008; Oswald & Golueke, 1966). The effects on environmental and human health, including decreases in potable and recreational water quality, loss of aquatic habitats, and disruption to food chains (Chorus & Welker, 2021; Erdner et al., 2008; Kozacek, 2014; Munn et al., 2018), are estimated to cost the country billions of dollars a year (Dodds et al., 2009).

Although reducing anthropogenic P inputs has been a focus of US policy for decades (Litke, 1999; USEPA, 1972), water quality improvements are often not timely or sufficient (Lintern et al., 2020; Sharpley et al., 2013; Stackpoole et al., 2019). While this is partially due to the lag time between the adoption of management practices and the detection of outcomes (Meals et al., 2010), it is also due to the continued difficulty in identifying and

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quantifying the vast number of persistent P sources and the release of legacy P from previous land management practices (Sharpley et al., 2013; Smith et al., 2019). As a result, the effectiveness of policies and other attempts to more sustainably manage P across the US, such as the improved management of both point and nonpoint sources and more extensive P recovery and recycling programs, has been relatively limited (Daneshgar et al., 2018; Haque, 2021; G. S. Metson et al., 2016).

The ability of phosphate (PO_4) dosed water to minimize lead and copper corrosion within water distribution networks has been understood for decades (Rice & Hatch, 1939; Schock, 1989). However, the contribution of this practice to the flux of P delivered to surface water environments around the world, via wastewater treatment plant (WWTP) effluents, has not been properly constrained, often due to lack of data which prevents the quantification of the relevant P fluxes (van Puijenbroek et al., 2019). PO_4 dosing of public water supply for corrosion control has been a widespread practice by US public water systems (PWS) since the passing of the Lead and Copper Rule in 1991 (McNeill & Edwards, 2002; National Research Council, 2006; Singley et al., 1984; USEPA, 1991). The importance of this dosing was recently highlighted by the Flint Water Crisis in 2014, where a lack of effective corrosion control practices resulted in multiple impacts, including the increased exposure of children to lead and a hypothesized range of associated long-term health effects (Edwards et al., 2009; Hanna-Attisha et al., 2015; Pieper et al., 2017).

Understanding the environmental impacts of PO_4 dosing regimes has been an active area of research largely focusing on the contribution of this dosing to influent loads of P at wastewater treatment plants (WWTPs) (USEPA, 2020a; Water Research Foundation, 2017). Beyond a limited number of small-scale studies (McNeill & Edwards, 2002; Rodgers, 2014), the extent of PO_4 dosing practices across the entire United States remains poorly constrained. In addition, not all PO_4 dosed water will be returned to WWTPs. Some PO_4 is thought to be released from the water distribution network and into the environment as a result of outdoor water use at domestic residences, the release of effluent from industrial cooling processes, and leakage of water from public water supply network pipes (USEPA, 2020a; Water Research Foundation, 2017). Despite this, research investigating these processes is lacking across the US.

Research has estimated the leakage flux of PO_4 dosed tap water from the water distribution network to the environment on both a national and catchment level across the UK (Ascott et al., 2016, 2018; Gooddy et al., 2017). During periods of high leakage, leakage fluxes of P were found to be equivalent to up to 20% of WWTP P inputs to rivers across urbanized catchments, highlighting their significance in these areas (Ascott et al., 2018). Leakage from distribution networks is ubiquitous within water systems around the world (Al-Washali et al., 2019; Lerner, 1990). With approximately 16% of the water entering the US watermains distribution network estimated to be lost due to leakage (USEPA, 2013), recent research has demonstrated that watermains leakage can be an important source of nitrogen fluxes to the environment across the United States (Flint et al., 2022). Therefore, we hypothesize that the leakage of dosed water may also constitute an important source of P across the country. Outdoor water use also represents a large proportion of total potable water use across many countries (Statistics Canada, 2017), with around one third of water supplied to domestic residences across the US being used outdoors each year (USEPA, 2017). As a result, water from public supply that has been used for lawn irrigation has been found to contribute significantly to baseflow across some US cities (Fillo et al., 2021) and PO_4 corrosion inhibitors have been reported as a potential source of P in urban runoff (Clary et al., 2020). Despite this, the fluxes of PO_4 to the environment that are associated with processes of leakage and outdoor water use are lacking, both in the United States and around the globe.

In this paper, we synthesize recommended PO_4 dosing concentrations (expressed as P), public water system dosing facility data, volumetric rate estimates of both public and domestic supply distribution inputs, as well as fractions of these inputs lost due to leakage and outdoor water use and use the resulting data set to address the following research questions:

1. What is the spatial variability of PO_4 dosing practices undertaken by public water systems for the purpose of corrosion control across US counties, and thus, what is the annual mass flux of PO_4-P added to these systems?
2. What is the annual mass flux of PO_4-P lost or actively removed from the watermains distribution network across US counties due to leakage and outdoor water use, respectively, and thus what is the residual mass flux returned to WWTPs per year?
3. What is the significance of, and dominant controls upon, estimated watermains leakage and outdoor water use PO_4-P fluxes across the United States?

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2. Materials and Methods

2.1. Estimating the Extent of PO_4 Dosing by Public Water Systems for Corrosion Control and the Mass Flux of PO_4-P Entering the Water Distribution Network

The mass flux of PO_4-P entering the water distribution network across each county of the contiguous United States (herein referred to as the US) due to dosing by public water systems (DOSE-PO_4-P_{pws}) was estimated using Equations 1–3. The fraction of a county's population served by public water systems that dose their water with either orthophosphate or polyphosphate (herein referred to as PO_4; f_{dosed}) was determined as the ratio of the county's population served by PO_4 dosed water (pop_{dosed}) to the total population served by public water systems (pop_{pws}). Pop_{dosed} values were determined through querying the Safe Drinking Water Information System (USEPA, 2022c) for public water systems (PWS) that had active PO_4 dosing facilities for the purpose of corrosion control in the year 2015. The resulting data set disclosed both the counties and the population size each PWS supplied, and corresponded with the most recent water use data release year (Dieter et al., 2018). County-level pop_{pws} values were sourced from Dieter et al. (2018).

$$f_{dosed} = pop_{dosed} / pop_{pws} \tag{1}$$

f_{dosed} was then applied to county-level volumetric rates of PWS distribution inputs to give the total volumetric rate of PO_4 dosed water entering the public water supply distribution network ($Vol_{pws\text{-}dosed}$) in L yr^{-1} (Equation 2). With PWS distribution input estimates omitted from water use reports, they were assumed to equal the total volume of freshwater withdrawn for public supply in each county for the year 2015 ($WD_{(pws\text{-}total)}$) in L yr^{-1}, and were sourced from Dieter et al. (2018).

$$Vol_{pws-dosed} = f_{dosed} \text{ x } WD_{(pws-total)} \tag{2}$$

DOSE-PO_4-P_{pws} values for each county, in kg PO_4-P yr^{-1}, were estimated as the product of $Vol_{pws\text{-}dosed}$ and PO_4-P concentrations within dosed tap water ($C_{t(PO4\text{-}P)}$), in mg L^{-1} (Equation 3). Due to generally low PO_4 concentrations in natural waters (Hem, 1985; Litke, 1999), and the lack of a health-based PO_4 limit within potable water (USEPA, 2021; World Health Organization, 2005), PO_4 concentrations are not widely reported for dosed or nondosed water. We therefore assumed that concentrations of PO_4-P in potable water were only present as a result of dosing practices by public water systems (D. Cornwell et al., 2015). Given no comprehensive national $C_{t(PO4\text{-}P)}$ data set, we adopted the USEPA (2016) recommended lower and upper target residual PO_4 concentrations at the consumers tap (0.33 and 1 mg L^{-1}, respectively) in order to make both lower and upper DOSE-PO_4-P_{pws} flux estimates. County-level DOSE-PO_4-P_{pws} estimates were aggregated to give a final national-level estimate, in metric kt PO_4-P yr^{-1}.

$$\text{DOSE-}PO_4\text{-}P_{pws} = Vol_{pws-dosed} \text{ x } C_{t(PO4-P)} \tag{3}$$

2.2. Estimating Watermains Leakage and Outdoor Water Use PO_4-P Fluxes, and Their Comparison With Other P Fluxes to the Environment

County-level PO_4-P fluxes due to watermains leakage (WML-PO_4-P) were estimated across the United States using Equations 4 and 5. $Vol_{pws\text{-}dosed}$ was adjusted using a leakage factor ($f_{leakage}$; unitless) to give a final volumetric rate of leaked dosed water ($Vol_{leaked\text{-}dosed}$) in L yr^{-1} (Equation 4). With the exception of California and Georgia, a lack of county-level leakage factor data meant that state level $f_{leakage}$ values were obtained from various sources (Table S1 and Figure S1a in Supporting Information S1) and assigned to their respective counties. For the 22 states without $f_{leakage}$ values, the national average of 0.16 was used (USEPA, 2013).

$$Vol_{leaked-dosed} = Vol_{pws-dosed} \text{ x } f_{leakage} \tag{4}$$

County-level WML-PO_4-P estimates, in kg PO_4-P yr^{-1}, were estimated as the product of $Vol_{leaked\text{-}dosed}$ and $C_{t(PO4\text{-}P)}$ (Equation 5). Effective corrosion control within in-building plumbing requires target $C_{t(PO4\text{-}P)}$ values to be met at the consumers tap. Due to PO_4 reacting with other compounds and influencing biological processes within pipe networks (Douterelo et al., 2020), water leaving dosing plants will often have higher PO_4 concentrations than those further along the network (Hill & Cantor, 2011; USEPA, 2016). Over time, dosing concentrations of PO_4 will equal those at the tap (Comber et al., 2013); however, the time to reach this equilibrium remains largely

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unknown and varies between individual water supply systems (USEPA, 2016). As a result, we assumed $C_{t(PO4\text{-}P)}$ values were the same along the entire pipe network. County-level WML-PO_4-P fluxes were aggregated to give a national-level estimate, in metric kt PO_4-P yr^{-1}, as well as normalized for land area, in kg PO_4-P km^{-2} yr^{-1}.

$$\text{WML-PO}_4\text{-P} = \text{Vol}_{\text{leaked-dosed}} \text{ x } C_{t(PO4-P)} \tag{5}$$

County-level fluxes of PO_4-P leaving the distribution network due to the use of PO_4 dosed water outdoors at domestic residences across the United States (OWU-PO_4-P) were estimated using Equations 6–8. The volume of PO_4 dosed water supplied for domestic use ($Vol_{domestic\text{-}dosed}$) was estimated as the product of public supply delivered to domestic users ($DI_{domestic}$) in L yr^{-1} reported by Dieter et al. (2018) and f_{dosed} (Equation 6). $Vol_{domestic\text{-}dosed}$ was corrected using an outdoor water use factor (f_{owu}; unitless) to give the volumetric flow rate of dosed water for outdoor use ($Vol_{owu\text{-}dosed}$) (Equation 7). The lack of county-level f_{owu} data meant that state-level values, ranging from 0.25 to 0.6, were obtained from various sources (Table S2 and Figure S1a in Supporting Information S1). For the 38 states without f_{owu} values, the national average of 0.3 was used (USEPA, 2017). County-level OWU-PO_4-P fluxes, in kg PO_4-P yr^{-1}, were estimated as the product of $Vol_{owu\text{-}dosed}$ and $C_{t(PO4\text{-}P)}$ (Equation 8) and were both normalized for land area, in kg PO_4-P km^{-2} yr^{-1} as well as aggregated to give a national-level estimate in metric kt PO_4-P yr^{-1}.

$$\text{Vol}_{\text{domestic-dosed}} = \text{DI}_{\text{domestic}} \text{ x } f_{\text{dosed}} \tag{6}$$

$$\text{Vol}_{\text{owu-dosed}} = \text{Vol}_{\text{domestic-dosed}} \text{ x } f_{\text{owu}} \tag{7}$$

$$\text{OWU-PO}_4\text{-P} = \text{Vol}_{\text{owu-dosed}} \text{ x } C_{t(PO4-P)} \tag{8}$$

Upper and lower estimates of national-level WML-PO_4-P and OWU-PO_4-P fluxes were determined by adjusting $C_{t(PO4\text{-}P)}$ to lower and upper target residual concentrations of 0.33 and 1.0 mg L^{-1} PO_4-P, respectively (USEPA, 2016), and $WD_{(pws\text{-}total)}$ and $DI_{domestic}$ values of ±10%, respectively. Although the inherent uncertainty associated with USGS withdrawal data is currently not reported (National Research Council, 2002), we have adopted ±10% uncertainty on $WD_{(pws\text{-}total)}$ and $DI_{domestic}$ values to reflect the uncertainty used within previous US water budget research (Maupin & Weakland, 2009).

County-level WML-PO_4-P and OWU-PO_4-P fluxes were summated and normalized for county area, in kg PO_4-P km^{-2} yr^{-1}. The significance of county and national-level WML-PO_4-P and OWU-PO_4-P fluxes, in the context of US P budgets, was evaluated through their comparison with estimates of P fluxes from other sources to the environment, including P fluxes to surface waterbodies from point sources, including both municipal and industrial WWTP effluents (USEPA, 2022b), the use of farm and nonfarm (urban) P fertilizers and P from manure application (Falcone, 2021). Key controls upon OWU-PO_4-P and WML-PO_4-P fluxes were investigated through their comparison with potentially influencing factors, such as population density.

County-level mass fluxes of PO_4-P returned to WWTPs as a result of PO_4 dosing were estimated using a mass balance equation (Equation 9 and Figure 1) and aggregated to give a national-level estimate, in metric kt PO_4-P yr^{-1}. It should be noted that while leaking and overflowing sewers and septic tanks have been found to be important sources of nutrient loading across the US (Delesantro et al., 2022; Iverson et al., 2018), estimating the loss of drinking water derived PO_4-P between domestic residences and WWTPs associated with these processes is beyond the scope of this research.

$$\text{DOSE-PO}_4\text{-P}_{\text{wwtp}} = \text{DOSE-PO}_4\text{-P}_{\text{pws}} - \text{WML-PO}_4\text{-P} - \text{OWU-PO}_4\text{-P} \tag{9}$$

3. Results

3.1. Estimating the Extent of PO_4 Dosing by Public Water Systems for Corrosion Control and the Mass Flux of PO_4-P Entering the Water Distribution Network

Our analyses reveal that, in 2015, 4,572 of the 152,104 active public water systems (PWS) across the US (3%) had at least one facility that dosed their water with PO_4 for the purpose of controlling lead and copper corrosion (Table 1 and Figure 2a). These facilities were found within 1,402 of the 3,109 US counties considered in our research (Figure 2b). The percentage of PWS that dose is positively correlated with the size of the PWS, as represented by the size category of population it serves (i.e., the number of people it serves) (Table 1). For example,

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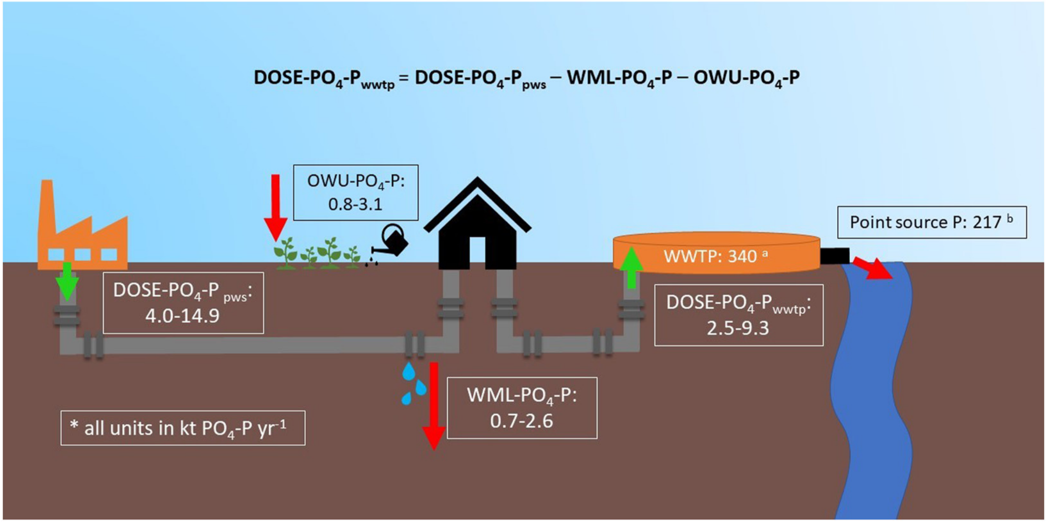

Figure 1. Schematic figure showing the national scale mass balance developed to estimate the contribution that PO_4 dosed water returned to wastewater treatment plants (WWTPs) (DOSE-PO_4-P_{wwtp}) makes to total estimated loading of P to municipal WWTPs for the year 2015. Flux ranges represent lower and upper-bound estimates for the input of PO_4 by public water systems (DOSE-PO_4-P_{pws}), losses from the water distribution network due to watermains leakage (WML-PO_4-P), outdoor water use (OWU-PO_4-P) and DOSE-PO_4-P_{wwtp}. These flux values may not sum due to rounding. [a] Hallas et al. (2019) and [b] USEPA (2022b).

25% of very large PWS (those that serve >100,000 people) dose water with PO_4 for corrosion control purposes, whereas the figure is only 1% for very small systems (that serve <500 people). However, when combined, the large absolute number of "small" and "very small" PWS (serving <500 and 501–3,300 people, respectively) means that they contribute over 60% of the total 4,572 PWS that dose with PO_4. Although the absolute number of PWS undertaking PO_4 dosing decreases with increasing PWS size catagory, the total population served by PWS that dose is also positively correlated with PWS size (Table 1). As a consequence, although only 3% of PWS across the United States dosed in 2015, 25% of the total population were served by PO_4 dosed water (Table 1). The number of PWS with PO_4 dosing facilities, as well as total and percentage of state populations they serve, are generally larger within constituent counties of Midwestern, Northeastern and Mid-Atlantic states (e.g., Minnesota, Illinois, New York, New Jersey, and Pennsylvania), as well as a number of Californian counties (Figure 2). Nationally, it was estimated that 4–14.9 kt PO_4-P yr^{-1} entered the distribution network as a result of PO_4 dosing (Figure 1 and Table S3 in Supporting Information S1).

Table 1
The Total Number of Public Water Systems (PWS) and the Population These Systems Served Across the United States, as Well as the Number of These Systems That Undertook PO_4 Dosing and the Population Served by PO_4 Dosed Water, for Each Defined PWS Size Category in the Year 2015

PWS size (by population served)	Total number of PWS[a]	Total population served[a]	Number of PWS that dose for corrosion control[b]	Population served by PWS that dose[b]	% of PWS that dose[c]	% of population served by PWS that dose[c]
Very small (<500)	124,291	13,913,830	1,316	290,260	1.0 (29)	2.0 (0.40)
Small (501–3,300)	18,487	24,255,378	1,572	2,358,574	8.5 (34)	10 (3.0)
Medium (3,301–10,000)	5,090	29,613,444	777	4,569,572	15 (17)	15 (6.0)
Large (10,000–100,000)	3,813	108,954,823	802	23,549,372	21 (18)	22 (30)
Very large (>100,000)	422	139,224,370	105	47,436,133	25 (2.0)	34 (61)
PWS (all)	152,104	315,961,845	4,572	78,203,911	3	25

Note. The percentage of PWS that undertook PO_4 dosing when compared to the total number of PWS, as well as the percentage of the population served by PO_4 dosed water when compared to the total population served by PWS are also shown for each PWS size category.
[a]USEPA (2022a). [b]USEPA (2022c). [c]Values in parentheses show the percentage contribution that the number of dosing public water systems and the populations they serve in each system size category makes to the total number of dosing public water systems and total population served by dosed water (n = 4,572 and n = 78,203,911, respectively).

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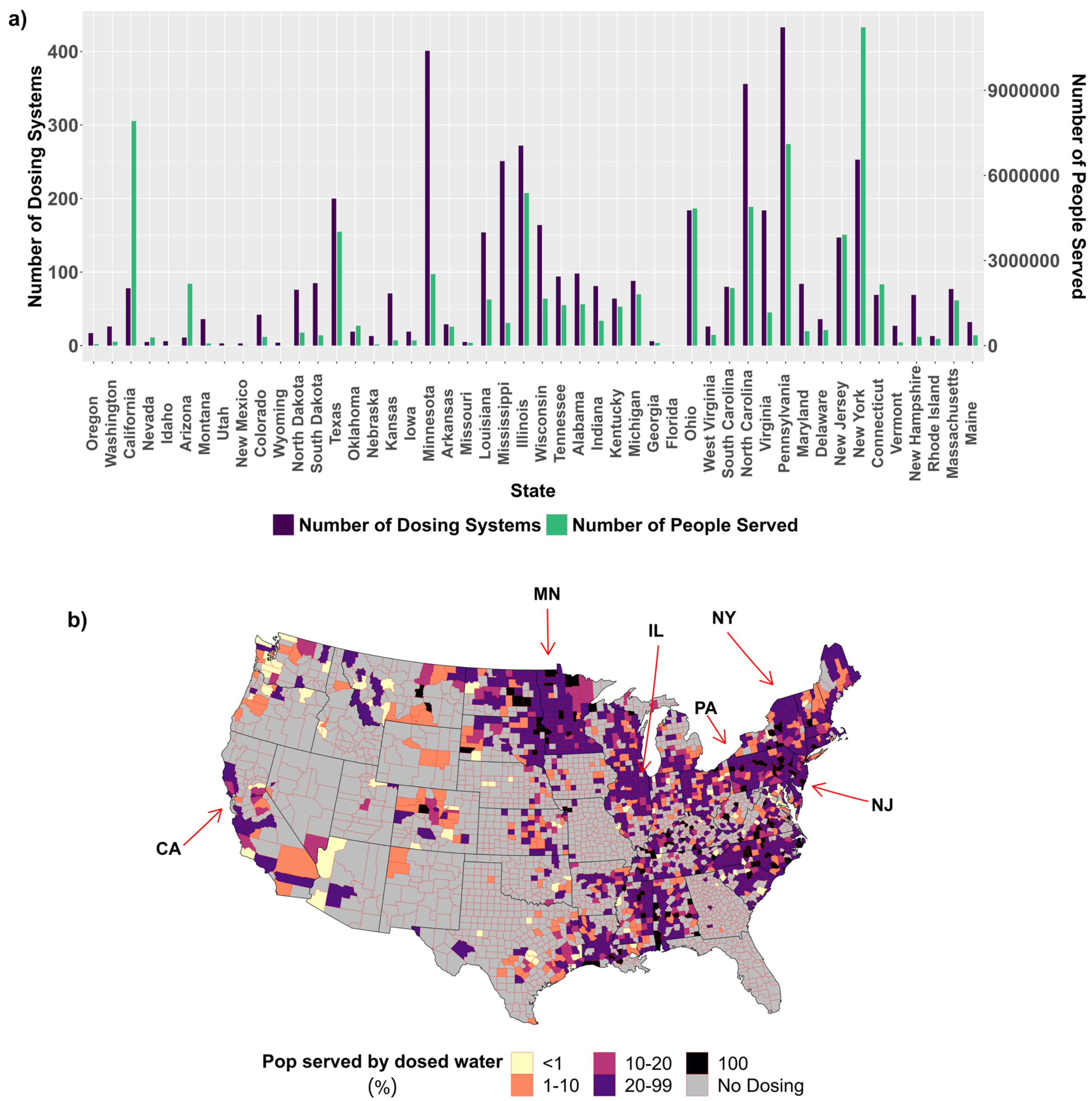

Figure 2. (a) The number of public water systems with active phosphate (PO_4) dosing facilities across the United States in the year 2015, displayed by purple bars, and the total population that these public water systems serve, displayed by green bars. States on the *x*-axis are ordered from west to east. (b) Percentage of the population supplied with PO_4 dosed water for each county across the United States, with state labels indicating areas with particularly high values. Linework was created using the "usmap" package in R (Di Lorenzo, 2022).

3.2. Watermains Leakage and Outdoor Water Use PO_4-P Fluxes

Nationally, an estimated 5%–17% of PO_4 dosed water within the water distribution network was lost due to watermains leakage, with the associated flux of PO_4-P (WML-PO_4-P) estimated to be between 0.7 and 2.6 kt PO_4-P yr^{-1}. Of the 1,402 counties with at least one PWS that undertook PO_4 dosing in 2015, 58% are defined as urban. A general trend of increasing WML-PO_4-P fluxes from west to east across the United States prevails, with the highest estimated fluxes observed in urbanized counties of Midwestern, Northeastern and mid-Atlantic states, such as Philadelphia County in Pennsylvania and Union County in New Jersey (Figure 3b).

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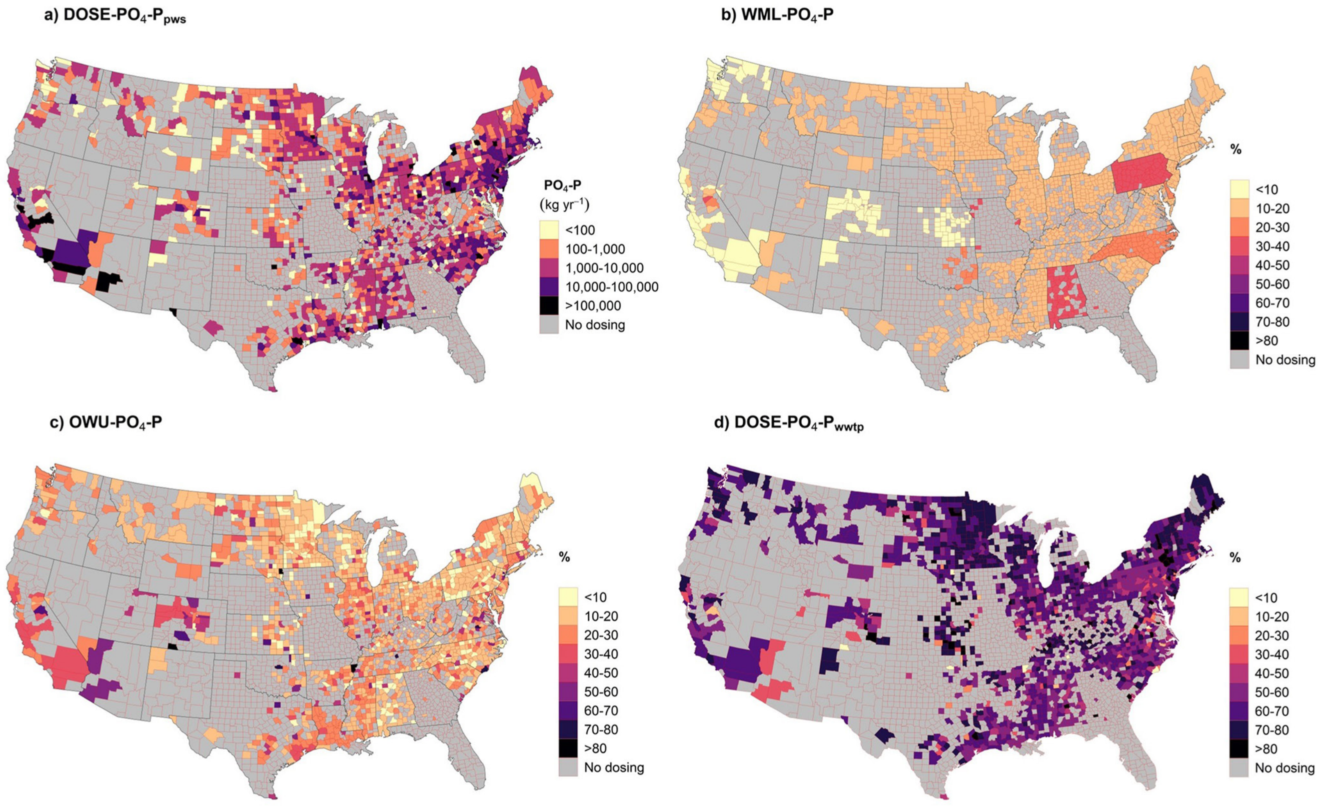

Figure 3. (a) The mass flux of PO_4-P added to public water supply distribution networks across each US county due to PO_4 dosing (DOSE-PO_4-P_{pws}), in units of kg PO_4-P yr^{-1}. The percentage of each county's the estimated DOSE-PO_4-P_{pws} flux that was lost from its water distribution network due to (b) watermains leakage (WML-PO_4-P) and removed due to (c) outdoor water use (OWU-PO_4-P). (d) The percentage of a county's DOSE-PO_4-P_{pws} flux returned to wastewater treatment plants (DOSE-PO_4-P_{wwtp}), once WML-PO_4-P and OWU-PO_4-P fluxes had been accounted for. All fluxes are for the year 2015, and gray areas indicate counties where no PO_4 dosing was reported. Linework was created using the "usmap" package on R (Di Lorenzo, 2022).

On a national-level, 5%–21% of PO_4 dosed water was removed from the water distribution network due to outdoor water use at domestic residences, with the associated PO_4-P flux (OWU-PO_4-P) estimated to be between 0.8 and 3.1 kt PO_4-P yr^{-1} (Figure 1 and Table S2 in Supporting Information S1). Counties in the Northeast and state of California had the largest OWU-PO_4-P fluxes (Figure 3c). When combined, the upper bound national WML-PO_4-P and OWU-PO_4-P flux estimates (2.6 and 3.1 kt PO_4-P yr^{-1}, respectively) are equivalent to around 12% of P inputs to the environment from urban fertilizer, 2.6% of the P load to surface waterbodies from point sources, and 0.3% of P inputs to the environment from farm fertilizers and manure application (Table 2).There is large intercounty variability in combined area-normalized WML-PO_4-P and OWU-PO_4-P fluxes, with estimates ranging from 0 to 817 kg PO_4-P km^{-2} yr^{-1} (Figure 4a). A strong linear relationship was observed between the combination of area-normalized OWU-PO_4-P and WML-PO_4-P fluxes and population density (r = 0.87, $p < 0.01$; Figure 5). When combined, lower and upper county-level WML-PO_4-P and OWU-PO_4-P flux estimates

Table 2
WML-PO_4-P and OWU-PO_4-P Fluxes as a % Equivalence of Other Estimates of Major P Inputs to the Environment

Flux	Urban P fertilizer input (47)[a]	Loads of P from point sources (217)[b]	Farm P fertilizer input (1,829)[a]	Manure P input (1,908)[a]
WML-PO_4-P (0.7–2.6)	1.5–5.5	0.32–1.2	0.038–0.14	0.037–0.13
OWU-PO_4-P (0.8–3.1)	1.7–6.5	0.37–1.4	0.044–0.17	0.042–0.16
WML-PO_4-P + OWU-PO_4-P (1.5–5.6)	3.2–12	0.69–2.6	0.082–0.30	0.079–0.30

Note. Values in parentheses are in units of kt PO_4-P yr^{-1}.
[a]Falcone (2021). [b]USEPA (2022b).

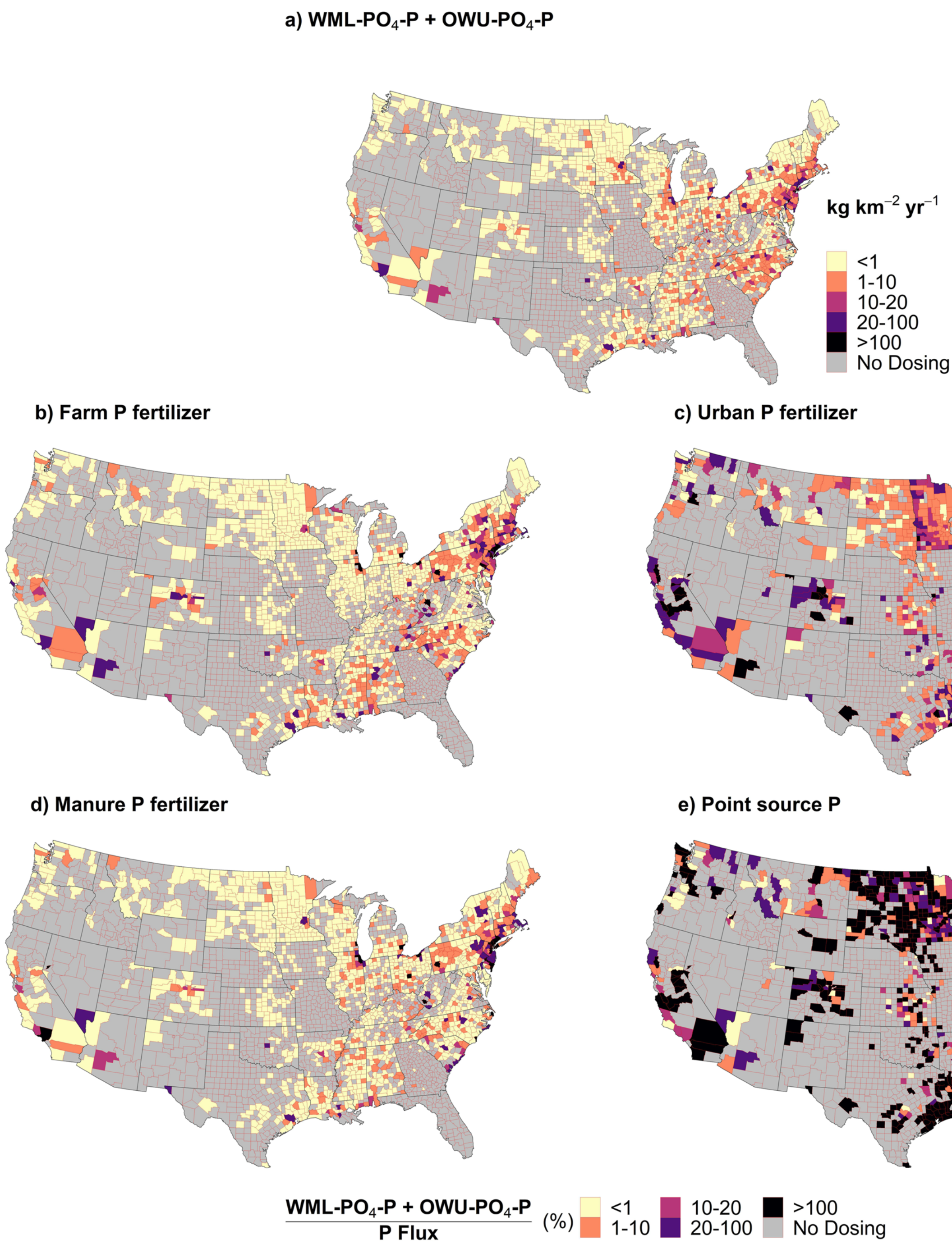

Figure 4. (a) The sum of estimated area-normalized county-level WML-PO_4-P and OWU-PO_4-P fluxes across the US for the year 2015; the sum of estimated county-level WML-PO_4-P and OWU-PO_4-P fluxes as a percentage of P fluxes from (b) farm and (c) urban fertilizer input (d) manure (e) P loads to surface waterbodies from point sources. Gray areas indicate counties where no PO_4 dosing was reported. Linework was created using the "usmap" package on R (Di Lorenzo, 2022).

exceed P inputs to the environment from urban and farm fertilizer usage and manure application across 16–56, 13–21, and 17–32 counties, respectively, and exceed P inputs from point sources to freshwaters across 461–541 counties, out of a total of 3,101 US counties analyzed in this research (Figures 4b–4e). In addition, when upper bound county-level estimates are considered, 39 counties have combined WML-PO_4-P and OWU-PO_4-P fluxes that exceed the sum of all major P inputs to the environment (farm and nonfarm fertilizer, manure and point source P inputs).

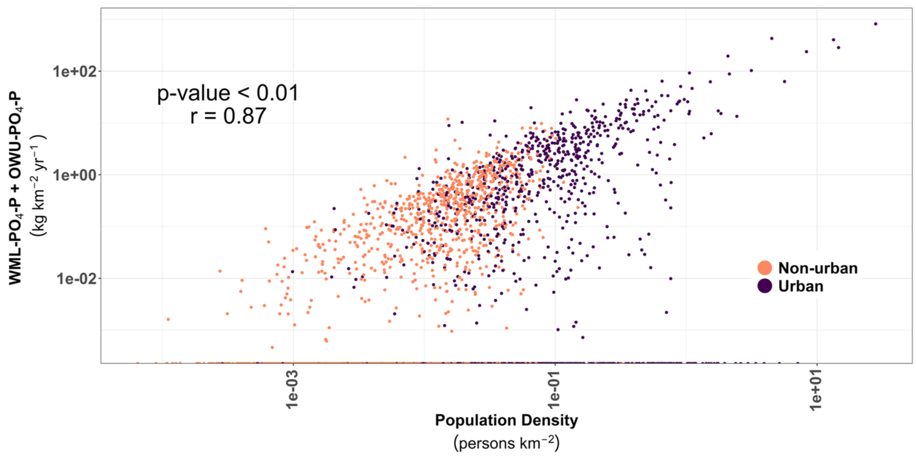

Figure 5. Relationship between the sum of combined area-normalized WML-PO_4-P and OWU-PO_4-P fluxes and population density (p-value < 0.01, $r = 0.87$) for both urban and nonurban counties across the US, as defined by the US Department of Agriculture. Counties with WML-PO_4-P and OWU-PO_4-P fluxes = 0 are where there is no P dosing.

The removal of lower or upper level national-level OWU-PO_4-P and WML-PO_4-P flux estimates from either the lower or upper bound national-level DOSE-PO_4-P_{pws} flux resulted in a total mass flux of P returned to US WWTPs as a result of PO_4 dosing (DOSE-PO_4-P_{wwtp}) estimated to be 2.5–9.3 kt PO_4-P yr^{-1} (Figure 1). Counties with the highest DOSE-PO_4-P_{pws} fluxes were found across the Northeast, mid-Atlantic and the state of California (Figure 3d).

4. Discussion

4.1. Controls on the Extent of PO_4 Dosing for Corrosion Control and Mass Flux of P Entering the US Water Distribution Network

The methodology and the data reported in this research have allowed us to report the first estimates of P fluxes associated with PO_4 dosing by public water systems (DOSE-PO_4-P_{pws}), watermains leakage (WML-PO_4-P), domestic outdoor water use (OWU-PO_4-P) and the returns of dosed PO_4-P to WWTPs (DOSE-PO_4-P_{wwtp}) at the scale of the contiguous United States. We show that DOSE-PO_4-P_{pws} fluxes across the United States may have been up to 14.9 kt PO_4-P yr^{-1}, in 2015, with up to 17% of this PO_4-P lost to the environment via watermains leakage and 21% input to the environment via domestic outdoor water use. Some upper bound county-level WML-PO_4-P and OWU-PO_4-P fluxes exceeded other well-constrained P fluxes to the environment, such as point source inputs. Once WML-PO_4-P and OWU-PO_4-P fluxes had been accounted for, we estimated the national DOSE-PO_4-P_{wwtp} flux to be up to 9.3 kt PO_4-P yr^{-1}.

The most recent and comprehensive analysis of the extent of PO_4 dosing, undertaken by the USEPA (2020a), estimates that the number of public water systems (PWS) undertaking dosing nationally is slightly larger than the one we present here (around 8,500, or 5.6%), suggesting that our approach may have underestimated the number of PWS undertaking dosing, and thus DOSE-PO_4-P_{pws}, WML-PO_4-P and OWU-PO_4-P estimates. Despite this, the analysis presented here provides the first subnational scale insight into the variance of PO_4 dosing across the country (Figure 3a).

The inverse relationship between the number of PWS that undertake PO_4 dosing and the size of population they serve (Table 1) is consistent with findings reported by McNeill and Edwards (2002). However, we find the proportion of PWS dosing to be lower for all PWS size categories when compared to previously published research (Arnold et al., 2019; McNeill & Edwards, 2002). While these previous studies investigated the extent of PO_4 dosing across the United States (concluding that more than 50% of utilities use PO_4 based corrosion inhibitors), they targeted a limited number of utilities (264 and 60 out of around 50,000, respectively). The disparity between their estimates, the USEPA (2020a) estimate (5.6%), and the one we report here (3%) may also be due to the bias incorporated within past research through only investigating medium to large size utilities, with our analysis suggesting that larger size PWS are more likely to undertake PO_4 dosing.

Although around 7% of the total US population is thought to be served by PWS with lead watermains pipes (D. A. Cornwell et al., 2016), our analysis suggests 26% of people served by community water systems were supplied with PO_4 dosed water. This disparity may be due to precautionary PO_4 dosing by PWS, given the lack of comprehensive lead water main pipe inventories (USEPA, 2019), alongside the fact that many remaining lead solder components are located within property boundaries, and are thus not the responsibility of the utility. PWS may also undertake PO_4 dosing in order to prevent other metals (copper, manganese, and iron) found within nonlead pipes to be released into the water distribution network (Comber et al., 2011; Lytle & White, 2014; Lytle et al., 2018; McNeill & Edwards, 2002; USEPA, 2016). Recent Lead and Copper Rule revisions may drive changes in both the spatial extent of PO_4 dosing practices and the PO_4 concentrations required in the future (USEPA, 2019, 2020a). Estimating the extent of future PO_4 dosing practices across the country, and the effect this might have upon WML-PO_4-P, OWU-PO_4-P, and DOSE-PO_4-P_{wwtp} fluxes, should be a priority for future research.

Urbanized counties across Midwestern and Northeastern regions, such as Philadelphia, Chicago, and Milwaukee, have the largest total and proportional populations served by dosed water (Figure 2). The higher proportion of PWS undertaking PO_4 dosing in these areas likely reflects the dense network of lead watermains pipes in these areas (D. A. Cornwell et al., 2016; NRDC, 2022) that would have been installed prior to the lead piping ban in 1986 (AWWA, 2012; USEPA, 1989). Higher dosing rates across many urbanized areas have also been linked to the elevated corrosivity of their raw surface waters (Stets et al., 2018). Further, corrosivity of raw groundwater used for public supply is also higher across eastern regions of the US, including the states of New Jersey, Maryland, Delaware, and South Carolina (Belitz et al., 2016), where higher dosing rates were observed (Figure 2). However, regions of the United States with a low prevalence of PO_4 dosing, such as Georgia (Figure 2), do not necessarily indicate a lower presence of lead watermains pipes because these areas use alternative corrosion control methods such as pH adjustment (USEPA, 2022c).

Prescribing a fixed lower or upper PO_4-P dosing concentration within our calculations (0.33 or 1.0 mg L^{-1} PO_4-P, respectively) will have propagated uncertainty to DOSE-PO_4-P_{pws}, WML-PO_4-P and OWU-PO_4-P and DOSE-PO_4-P_{wwtp} estimates. In reality, PWS across the United States add PO_4 in varying concentrations both within and outside of the USEPA target range (Comber et al., 2013; The Cadmus Group Inc., 2004; USEPA, 2016). When PWS first establish PO_4 dosing regimes, they may add PO_4 at concentrations two to three times higher than the target concentration required at the consumers tap, meaning that county-level DOSE-PO_4-P_{pws} values may have been underestimated in the research reported here, at least for any PWS in the early stages of establishment. While assuming a dosing concentration of 3 mg L^{-1} PO_4-P across the entire United States would result in a DOSE-PO_4-P_{pws} flux of 44.7 kt PO_4-P yr^{-1}, opposed to the 14.9 kt PO_4-P yr^{-1} upper estimate reported in this research, these higher doses are often only needed to be applied for a few weeks before they can be reduced back to target maintenance concentrations (Hill & Cantor, 2011; MOE, 2009; USEPA, 2016). Along with the fact that concentrations of PO_4 can also vary with distance along the distribution pipe network, improving our understanding of PO_4-P concentrations at various points along water distribution networks is fundamental to better constrain the uncertainties associated with DOSE-PO_4-P_{pws}, WML-PO_4-P, OWU-PO_4-P, and DOSE-PO_4-P_{wwtp} fluxes.

4.2. Losses of PO_4 From Water Distribution Networks Due To Watermains Leakage and Outdoor Water Use and Implications for P Returns to Wastewater Treatment Plants

While a number of previous studies have assumed that residual PO_4-P loads within the distribution network will ultimately be returned to WWTPs (Comber et al., 2013; Vaccari, 2011), the research reported here highlights that these residual loads may also be released into the environment and bypass WWTPs. Our results suggest that 5%–17% and 5%–21% of DOSE-PO_4-P_{pws} are either lost from the water distribution network across the United States due to watermains leakages or removed due to outdoor water use at domestic residences, respectively. While the USEPA (2020a) incorporated leakage and outdoor water use within their conceptual mass balance model to investigate increases in P loading at WWTPs as a result of dosing for corrosion control, a single rate of water loss was applied to each process across the country. Further, the study did not report the national-level, or any subnational scale variance or significance of the P loss estimates as potential sources of P to the environment in their own right.

While the use of more locally determined leakage rates within this research, either on a utility-level for the states of California and Georgia or state-level where possible, allows for greater spatial resolution in WML-PO_4-P estimates when compared to previous research, our results still do not fully capture the localized variance in leakage rates across the country. The fraction of water lost due to leakage ($f_{leakage}$) can vary significantly even within a single state, with Californian utilities having leakage factors varying between <0.01 and 0.75 for the year 2015 (California Department of Water Resources, 2019; DeOreo et al., 2011). It is likely that many counties within the Northwest and Midwestern regions of the US will have locally elevated water leakage rates due to the aging condition of infrastructure in these areas (Folkman, 2018). However, the use of state-average factors will have masked these highly localized differences and therefore introduces further uncertainty into the resulting WML-PO_4-P flux estimates.

The largest county-level OWU-PO_4-P fluxes were observed across densely populated urban areas in the Northeastern US (Figure 3c). These are areas associated with the largest public supply deliveries to domestic users (Figure S1c in Supporting Information S1), although the fraction of water used outdoors (f_{owu}) at domestic residences is often below the national average (Figure S1a in Supporting Information S1). The effect of population density upon public supply withdrawals and domestic deliveries, and thus on both WML-PO_4-P and OWU-PO_4-P fluxes, is highlighted in Figure 5. Higher f_{owu} values across the Southwestern United States (Table S2 in Supporting Information S1), due to the arid climate in these areas (USEPA, 2017), contribute to the larger OWU-PO_4-P estimates across constituent counties within this region, such as California (Figure 3c). Despite this, the use of national f_{owu} values masks the large difference in outdoor water use rates between and within single states (DeOreo et al., 2011), as well as individual cities (Mini et al., 2014), thus adding uncertainty to OWU-PO_4-P flux estimates. Future work should aim to utilize more localized $f_{leakage}$ and f_{owu} values, as well as to incorporate seasonality in both leakage (Folkman, 2018; Healey et al., 2021) and outdoor water use rates (Opalinski et al., 2020), to enhance the accuracy of WML-PO_4-P and OWU-PO_4-P estimates.

While we assume that PO_4 will only be present within publicly supplied water if added for corrosion control purposes, low PO_4 concentrations are also found within nondosed water. This may be associated with both natural processes and other human activities, for example, resulting in median concentrations of nondosed groundwater used for US public supply reaching 0.033 mg L^{-1} PO_4-P (Hem, 1985; Kent et al., 2020). With ubiquitous watermains leakage and outdoor water use across all water systems (USEPA, 2017), low-level WML-PO_4-P and OWU-PO_4-P fluxes will occur even in the absence of dosing and are likely to have resulted in somewhat conservative WML-PO_4-P and OWU-PO_4-P flux estimates.

As a result of the data limitations described above, hotspots of WML-PO_4-P and OWU-PO_4-P fluxes are largely determined by differences in public supply withdrawals and domestic water deliveries, respectively (Figures S1b and S1c in Supporting Information S1). Higher county-level WML-PO_4-P fluxes, particularly across Midwestern and Eastern regions (Figure 3b), not only reflect the higher proportion of PO_4 dosing in these areas (see Section 4.1) but also larger $WD_{(pws\text{-}total)}$ and $f_{leakage}$ values in these areas (Figure S1 and Table S1 in Supporting Information S1). For example, densely populated urban areas, such as the cities of New York, Chicago and Philadelphia, are underlain by dense networks of water main pipes (Bonneau et al., 2017) that are capable of supplying higher volumetric rates of water for public supply, as reflected in county-level water use estimates made by Dieter et al. (2018) (Figures S1b and S1c in Supporting Information S1).

Once WML-PO_4-P and OWU-PO_4-P fluxes had been accounted for, our national-scale DOSE-PO_4-P_{wwtp} estimate (9.3 kt PO_4-P yr^{-1}) was in broad agreement with the 6 kt PO_4-P yr^{-1} reported by the USEPA (2020a), thereby supporting the robustness of the method developed in our research. This annual DOSE-PO_4-P_{wwtp} flux is equivalent to approximately 2.7% of the total inflow P load to municipal WWTPs estimated by Hallas et al. (2019) and is relatively low when compared to other major contributors, such as human excreta and detergents (Vaccari, 2011). In proportional terms, this is below the 6% estimated for WWTPs in England (Comber et al., 2013). This likely reflects the lower prevalence of PO_4 dosing across the United States compared to the UK, with around 95% of water supplies in the UK being dosed (CIWEM, 2011; Environment Agency, 2019) compared to the 25% we estimated across the United States. The nonubiquitous nature of dosing across the United States means that in the 84% of counties where dosing is undertaken, PO_4 dosed water may represent a larger proportion of the P loads entering their constituent WWTPs than suggested by the national-scale figure (Rodgers, 2014). Assessing the proportional contribution that DOSE-PO_4-P_{wwtp} fluxes make to individual WWTP influent P loads is beyond the scope of this study, although is an important area for future research.

Leaking and overflowing sewage infrastructure and septic tanks are commonplace across the United States (ASCE, 2017), and can be major sources of nutrients to catchments across the country (Delesantro et al., 2022;

Iverson et al., 2018). Omitting losses of drinking water derived PO_4-P from sewage infrastructure due to these processes within the approach used here to estimate DOSE-PO_4-P_{wwtp} (Equation 9) may have led to overestimation of DOSE-PO_4-P_{wwtp} values. Improved understanding and quantification of P losses from sewage infrastructure should also be a future research priority.

The localized significance of WML-PO_4-P and OWU-PO_4-P fluxes (Figure 3) highlights that these should be included within WWTP mass balances that aim to quantify P loads and financial impacts associated with PO_4 dosing practices (The Cadmus Group Inc., 2004). A nation-wide analysis of these loads to individual WWTPs is imperative, as P treatment is estimated to cost WWTPs around $2.08 per kg of P incrementally added upstream at drinking water treatment plants prior to distribution (USEPA, 2020a). Further, these mass balances would also reveal the extent to which P dosing ultimately contributes to the release of P into the environment from WWTPs.

4.3. Significance and Environmental Impacts of Watermains Leakage and Outdoor Water Use PO_4-P Fluxes

WML-PO_4-P and OWU-PO_4-P fluxes represent not only a loss of water and P from distribution networks but also an additional source of P to the environment. Unlike the fluxes of nitrate nitrogen associated with watermains leakage, where leakage acts to return nitrate that was previously retained via public supply water withdrawals (Flint et al., 2022), WML-PO_4-P fluxes represent a new source of P to the local environment. On a national-level, WML-PO_4-P and OWU-PO_4-P fluxes were small when compared to other major P inputs (Table 2). The inclusion of these fluxes within US national-level P source apportionment studies, as they have been in the UK (Environment Agency, 2019; Gooddy et al., 2017), could support more informed P source management strategies (Sabo et al., 2021; Smith et al., 2019). The exceedance of county-level WML-PO_4-P and OWU-PO_4-P fluxes over other major P sources (Figure 4) supports calls for more localized nutrient management approaches that consider these WML-PO_4-P and OWU-PO_4-P fluxes when developing best management practices for individual watersheds (Frei et al., 2021; Hejna & Cutright, 2021; Mooney et al., 2020; Smith et al., 2019).

Climate change and an increasing population are growing threats to the quality and availability of drinking water across the United States (Brown et al., 2019), and concerns surrounding watermains leakage and outdoor water as unsustainable uses of water and energy are rising globally (Chini & Stillwell, 2018; Gober et al., 2016; Jarvie et al., 2015; Xu et al., 2014). Watermains leakage reduction and more conservative outdoor water use are established tools for more sustainable water management (Mini et al., 2014; Rupiper et al., 2022). However, the WML-PO_4-P and OWU-PO_4-P fluxes we report now highlight the additional need to integrate P and water resource management strategies (G. S. Metson et al., 2012).

Although the fate of leakages from watermains is largely unknown (D'Aniello et al., 2021), most leakages are not major burst events that are visible from the surface, but instead occur into the subsurface and go relatively undetected (Rupiper et al., 2022). We hypothesize that WML-PO_4-P fluxes will largely be released into the shallow subsurface alongside watermains leakage nitrate fluxes (Flint et al., 2022), similar to leaking sewage mains (Howard & Gerber, 2018; Lee et al., 2015; Pennino et al., 2016; Sercu et al., 2011). Once released, these fluxes may be transported through the vadose zone and within groundwater flow to surface water environments via base flow (Fillo et al., 2021), potentially contributing to elevated concentrations of P in surface waters (Howard & Gerber, 2018). The fate of WML-PO_4-P fluxes will depend on local watershed hydrology and a range of widely varying environmental conditions. For example, soils rich in calcium carbonate, clays and metal oxides are more likely than sandy soils to reduce the movement of PO_4 due to adsorption (Domagalski & Johnson, 2012; Smith et al., 2019). This temporary retention of leakage-derived PO_4 may then contribute to legacy P that may be released in the future, thus hampering the water quality response time of the future of P management practices (Sharpley et al., 2013). Pipe infrastructure can also alter hydrology, such as through the creation of subsurface fractures, meaning that urbanization may not only influence the magnitude of WML-PO_4-P and OWU-PO_4-P fluxes (Figure 5) but also enhance the transport and alter the fate of these fluxes within the shallow subsurface (Bonneau et al., 2017; Howard & Gerber, 2018; Kaushal & Belt, 2012).

Around half of the water used outdoors across the United States is for lawn irrigation. Inefficient watering practices across the country mean that a proportion of this water will leach into the subsurface or be lost via runoff (USEPA, 2017). Quantifying the amount of drinking water derived PO_4 that is being applied to lawns will allow us to estimate the extent to which this P already contributes to fertilization requirements, and thus help to address the unsustainable use of P-based fertilizers across many urban watersheds (Hobbie et al., 2017).

The PWS responsible for DOSE-PO_4-P_{pws}, WML-PO_4-P, and OWU-PO_4-P fluxes, and the downstream WWTPs and agencies responsible for P management, may extend beyond county, watershed and country boundaries. For this reason, understanding the fate of both OWU-PO_4-P and WML-PO_4-P fluxes and the potential for these fluxes to modify DOSE-PO_4-P_{wwtp} fluxes and contribute to elevated P concentrations within freshwater environments remains a critical area of future research. The characterization of the stable oxygen isotope composition of PO_4 dosed public supply water may provide an important framework and isotopic label through which to explore the fate of these fluxes (Davies et al., 2014; Gooddy et al., 2015).

The transferrable methodology developed in this research could help quantify fluxes of PO_4-P associated with leakage and outdoor water use in other locations that use PO_4-P based corrosion inhibitors. Minimizing OWU-PO_4-P and WML-PO_4-P fluxes is important for reducing the reliance of water and wastewater industries upon sparse and finite P rock reserves. Additionally, reducing these fluxes will increase DOSE-PO_4-P_{wwtp} fluxes, and thus enhance the potential for P recovery and recycling at WWTPs, a key requisite for more sustainable P use (Haque, 2021).

5. Conclusions

We estimate that PO_4 dosing of publicly supplied water for corrosion control purposes across the United States added 4–14.9 kt PO_4-P yr^{-1} into the water distribution network in 2015. We estimate that watermains leakage and outdoor water use resulted in 5%–17% and 5%–21% of this added PO_4-P (0.7–2.6 and 0.8–3.1 kt PO_4-P yr^{-1}) to be lost or actively removed from the water distribution network, respectively. These estimates suggest that up to 9.3 kt PO_4-P yr^{-1} of the PO_4 initially added was returned to WWTPs, representing around 2.7% of the national WWTP influent P load estimated for the United States. We demonstrate that county-level PO_4 dosing, watermains leakage and outdoor water use PO_4-P fluxes were heterogenous across the United States. The greater prevalence of PO_4 dosing across urbanized counties in Midwestern and Eastern regions of the country likely reflects the presence of legacy lead piping in these areas. When combined, regions with the largest area-normalized watermains leakage and outdoor water use PO_4-P fluxes were also found across these same regions. This reflects not only the occurrence of PO_4 dosing but also the larger volumes of water required to supply more dense populations in these areas. Estimates reported in this paper represent an initial assessment of the significance of dosing-derived PO_4-P fluxes in the context of existing US P budgets, with lower and upper estimates of watermains leakage and outdoor water use PO_4-P fluxes exceeding P loads to surface waterbodies from point sources across 461–541 counties. Future work should seek to use the methodology developed in this paper with utility specific data to generate more accurate estimates of these fluxes. The significance of these fluxes in the context of other major anthropogenic P inputs encourages their inclusion within P source apportionment studies and could help develop more effective P management strategies, particularly within urban areas, both in the United States and more widely around the world.

Data Availability Statement

All data used within this research are publicly available. Volumetric rate data of public water supply withdrawals and domestic water inputs were sourced from Dieter et al. (2018), target PO_4 dosing values were sourced from the USEPA (2022c) and outdoor water use and leakage factors were available from a variety of sources, see Supporting Information S1. State-level DOSE-PO_4-P_{pws}, OWU-PO_4-P, and WML-PO_4-P fluxes for 2015 are detailed in Supporting Information S1. Linework on map figures were created using the "usmap" package in R (Di Lorenzo, 2022), which is available under the GNU General Public License version 3.

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