Water Practice & Technology

Water Practice & Technology Vol 16 No 4, 1434 doi: 10.2166/wpt.2021.076

Temporal variations of *de facto* wastewater reuse and disinfection by-products in public water systems in the Shenandoah River watershed, USA

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ABSTRACT

Temporal variations of *de facto* wastewater reuse are relevant to public drinking water systems (PWSs) that obtain water from surface sources. Variations in wastewater discharge flows, streamflow, *de facto* reuse, and disinfection by-products (DBPs – trihalomethane-4 [THM4] and haloacetic acid-5 [HAA5]) over an 18-year period were examined at 11 PWSs in the Shenandoah River watershed, using more than 25,000 data records, in gaged and ungaged reaches. The relationship of *de facto* reuse with DBPs by year and quarter at the PWSs was examined. A linear relationship was found between THM4 and *de facto* reuse on an annual average basis ($p = 0.050$), as well as in quarters 3 (July – September) ($p = 0.032$) and 4 (October – December) ($p = 0.031$). Using a t-test ($p < 0.05$), the study also showed that there were significant differences in DBP levels for PWSs relative to 1% *de facto* reuse. This was found for THM4 based on annual average and quarter 1 (January – March) data, and for HAA5 based on quarter 3 data during the period of record.

Key words: DBP, *de facto* reuse, drinking water, trihalomethanes, water

HIGHLIGHTS

- *De facto* wastewater reuse demonstrates temporal variations.
- Wastewater related to use as an indicator of precursors for disinfection byproducts.
- *De facto* reuse related to trihalomethanes and haloacetic acids in drinking water water systems.
- Threshold value for *de facto* wastewater reuse and safe drinking water.
- Data to help inform decision-making about wastewater reuse to improve downstream water quality.

INTRODUCTION

De facto wastewater reuse is an important water cycle component in any given watershed. It has been shown to contribute a substantial portion of surface water flows across the United States (U.S.) and other countries (e.g., China, Germany, and the Netherlands), with higher levels during drought or other low streamflow conditions (Rice *et al.* 2013; Wang *et al.* 2017; Beard *et al.* 2019; Karakurt *et al.* 2019) and noted as needing further study in South Africa (Swana *et al.* 2020). *De facto* reuse based on average streamflow has been estimated as at least 1% for approximately 50% of U.S. drinking water treatment plant (DWTP) intakes (i.e., 1% was the median contribution of wastewater flow to U.S. DWTPs (Rice & Westerhoff 2015) and provides a realistic reference point that would be applicable to a broad range of such plants). Others have examined the impacts of *de facto* reuse on small communities (Nguyen & Westerhoff 2019) and found that DWTPs serving them have higher levels of *de facto* reuse.

The potential impact of *de facto* reuse on surface water quality and downstream DWTPs is increasingly being studied – for example for endocrine disrupting compounds and other chemicals of emerging concern (Rice *et al.* 2015; Ternes *et al.* 2015; Nguyen *et al.* 2018; Tran *et al.* 2018; Barber *et al.* 2019; Medlock-Kakaley *et al.* 2020),

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disinfection by-products (DBPs) (Rice *et al.* 2015; Weisman *et al.* 2019), and pathogens (Rice *et al.* 2015; Amoueyan *et al.* 2017; Caicedo *et al.* 2019; Soller *et al.* 2019). These studies showed a range of wastewater effluent impacts on downstream water quality.

De facto reuse based on the proportion of wastewater in any stream varies with time, as wastewater discharge flows and streamflow vary (Merritt 1983; Corbitt 1999; Fernandez *et al.* 2008; Daelman *et al.* 2015). Municipal wastewater discharge flows are generally considered directly related to the number of people served by a wastewater treatment plant (WWTP). Minimum design ranges for U.S. WWTPs have been reported as 0.95–1.5 cubic meters (m^3)/day/person (250–400 gallons (gals)/day/person) (Merritt 1983). WWTP flow is also influenced by infiltration and inflow (I&I) associated with heavy rain events. Many WWTPs also collect, treat, and discharge industrial wastewaters – that is, those discharged by industrial facilities including poultry plants, dairies and breweries.

Streamflow variations over time are mainly influenced by precipitation amount and intensity, and the watershed's characteristics, including drainage area, slope, land use, antecedent moisture conditions, and vegetation (Mays 2011). Climate change is related to increased levels of extreme weather events, including more severe droughts and more extreme precipitation events, and can contribute to increased streamflow variability (Sridhar *et al.* 2019). Evaporation, transpiration, groundwater recharge, stream channelization, and water abstraction also influence streamflow, in addition to wastewater discharges (Mays 2011).

De facto reuse may influence the levels and types of precursors associated with DBP formation in surface water-sourced public water supplies (PWSs; in the U.S., referred to as public water systems) (a PWS may contain more than one DWTP). Weisman *et al.* (2019), working in the Shenandoah River watershed, Virginia and West Virginia, USA, found that the concentrations of trihalomethane-4 (THM4) and haloacetic acid 5 (HAA5) increase in drinking water systems as *de facto* reuse increases in their source waters. (THM4 comprises the sum of concentrations of chloroform, bromodichloromethane, chlorodibromomethane, and bromoform, and is referred to in U.S. Environmental Protection Agency (USEPA) drinking water regulations as 'total trihalomethanes', while HAA5 comprises the summed concentrations of monochloroacetic, dichloroacetic, trichloroacetic, monobromoacetic, and dibromoacetic acids.) The study relied on WWTP discharge flows for a single year (i.e., 2015) and average streamflow over the period 1971–2000, and the assumption that the discharge rates and streamflows were constant, even though they were expected to vary within the study period.

While WWTPs remove a substantial fraction of the organic load prior to discharge, removal is not complete and some is discharged, including carbonaceous and nitrogenous compounds. When those waters are a PWS source, the residual load can serve as precursors and results in DBP formation. DBPs form during water treatment when chlorine is added to water containing organic and inorganic precursors to kill or inactivate microbiological pathogens (McGuire *et al.* 2014). DBP concentrations in water systems vary over time as a function of the amount and nature of the precursors present, and other factors such as the disinfectant type and dose, temperature (THM4 concentrations are typically higher in warmer months), pH (more THM4 is produced at higher pH levels), and time following chlorine addition (longer residence times are associated with higher THM4 concentrations) (Wang *et al.* 2017; Kennedy *et al.* 2021).

This study relates to variations in wastewater discharge flow, streamflow, and *de facto* reuse over 18 years in the Shenandoah River watershed. The resulting potential variations in the relationship of *de facto* reuse with DBPs (THM4 and HAA5) at PWSs in the watershed were also examined, and compared with the results from the prior study in which temporal variations were ignored (Weisman *et al.* 2019). The hypotheses tested, for annual average and quarterly conditions, were that DBP concentrations would be related to proportional *de facto* reuse and that a 1% threshold of such reuse (i.e., the median value for U.S. DWTPs) would have a significant impact on the level of DBP formation.

METHODS

In this study, WWTP discharges to and PWS withdrawals from surface waters in the Shenandoah River watershed were modeled over the period 2002–2019 (18 years). Variations in WWTP flow, streamflow, *de facto* reuse, and DBP concentrations (THM4 and HAA5) were examined. The Shenandoah River watershed was chosen because it has a relatively small group of PWSs that abstract supplies from its surface waters, the majority of which use chlorine, and are less likely to have confounding factors – for example, complex distribution systems and/or lengthy residence times – than other watersheds.

The Shenandoah watershed covers portions of northwestern Virginia and eastern West Virginia, in the mid-Atlantic portion of the USA, with three 8-digit hydrologic unit codes (HUCs) (Figure 1). It is the drinking water source for more than 200,000 people (U.S. Geological Survey (USGS) 2020a). *De facto* reuse (proportion of volumetric flow (%)) during the period was estimated at 11 surface water PWS intakes and evaluated during annual average and quarterly streamflow conditions (Quarter 1: January – March; 2: April – June; 3: July – September; 4: October – December). *De facto* reuse (%) was compared to PWS concentrations of THM4 and HAA5 (i.e., in the distribution systems) to evaluate its potential relationship to DBP formation in them and the potential impact of 1% *de facto* reuse on DBP formation. Statistical modeling was performed using R (R Core Team 2017).

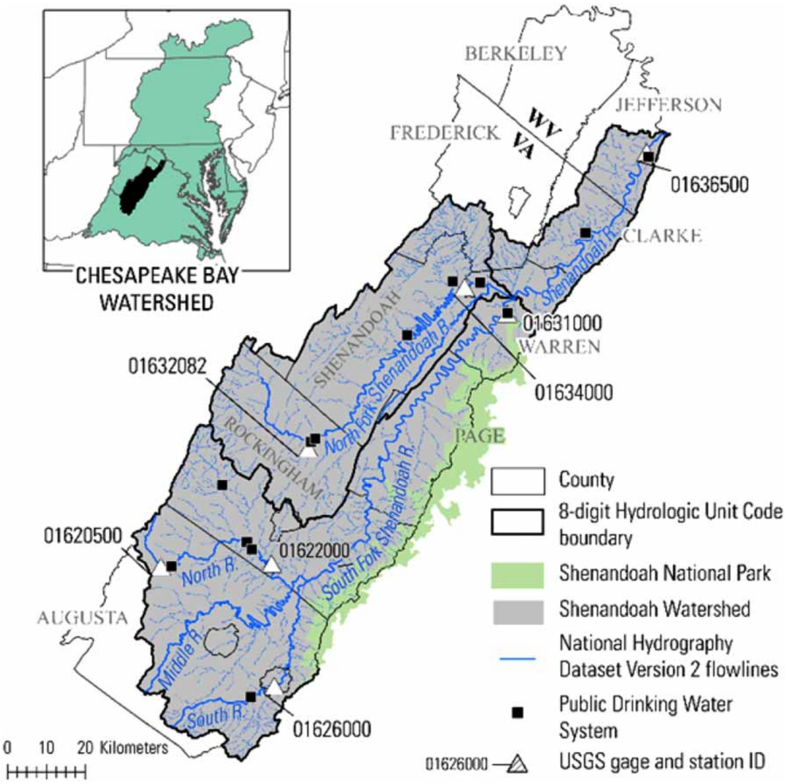

Figure 1 | The Shenandoah River watershed showing USGS stream gages (with identification numbers), PWSs, and National Hydrography Dataset Version 2 flowlines.

Flow data for WWTPs discharging into the Shenandoah watershed were obtained from discharge monitoring report databases compiled by state environmental departments for facilities with permits (Adams 2019; USEPA 2020). WWTP flows were compiled for the period from 2000 to end 2019 (DBP data were not available for 2000 and 2001).

For each WWTP, multiple outfall discharges in any month were combined to give a total monthly flow, and annual averages were calculated as the average of monthly values (only annual average data were collected for those WWTPs in West Virginia). WWTPs providing fewer than 10 years of flow data were ignored in the evaluation. The resulting dataset comprises 98 WWTPs – 22 industrial and 76 municipal WWTPs (94 in Virginia and four in West Virginia). WWTP flow and treatment information were verified by oral communication with the plant manager or their designee (2020) for nine of the 14 WWTPs that had calculated annual average flows exceeding or equal to 0.044 m^3/second (sec)] (1 million gals per day [MGD]). These larger WWTPs were upgraded to enhanced nutrient removal around the mid-point of the study period. Annual average WWTP flows in the watershed were calculated by year from the average flows from the 98 WWTPs.

The 11 surface water PWSs in the Shenandoah watershed comprise all the surface-water-supplied community systems in the watershed using conventional treatment (i.e., coagulation/flocculation, sedimentation, and filtration). Five of them serve populations of 10,000–100,000 people, the remainder serve smaller populations – the average and median populations served are 15,800 and 6,100, respectively. Ten systems use chlorine for disinfection, the other chloramine. Equally, six use a single surface water source each, the remaining five use either multiple surface water sources or a combination of surface- and groundwater. In addition to the above, it is understood that all 11 PWSs maintain relatively similar chlorine levels in their distribution systems and have residence times generally in the range of 1–2 days (oral communication with the plant manager or their designee (2017)), suggesting likely consistency in DBPs at monitoring points. The bromide concentration was below the minimum reporting level (0.02 milligrams per liter (mg/L)) in the source water for all five systems serving populations exceeding 10,000 people (USEPA 2021).

De facto reuse was estimated for the PWS intakes using geospatial modeling, to estimate wastewater accumulation in a river reach based on effluent discharge from municipal and industrial WWTPs and streamflow (Barber *et al.* 2019; Kandel *et al.* 2019; USGS 2019). Model inputs included reported WWTP discharge flows (Adams 2019; USEPA 2020), National Hydrography Dataset Plus Version 2 (NHDPlus V2) stream networks and hydrologic attributes (USEPA 2012), and measured streamflow discharge data from USGS continuous monitoring streamflow-gaging stations (USGS 2020b). A Python script (Python Core Team 2015) was used to summarize wastewater inputs by river segment common identifier (COMID) and calculate an accumulated wastewater (ACCWW) flow for each COMID, representing the total wastewater discharged upstream of the reach of interest. The intake ACCWW was used to estimate *de facto* reuse expressed as a percentage of total streamflow (the sum of the stream discharge and ACCWW) (Equation (1)),

$$\textit{de facto}\ \text{reuse} = \left[\left(\frac{\text{ACCWW}}{\text{Streamflow} + \text{ACCWW}}\right)\right] \times 100 \tag{1}$$

where streamflow represents stream discharge over a period of interest. For each intake, historic stream discharge values from the nearest upstream or downstream streamflow-gaging station were used to calculate mean and median *de facto* reuse for each year or quarter for the study period. For most intakes, stream gage measured flow was adjusted to represent river conditions at the intake before calculating the ACCWW. The adjustment factor was determined by the proportional difference in the estimated NHDPlus V2 mean annual streamflow conditions between the streamflow gage and intake river segments, and the gage's adjusted quarterly flow calculated as the product of its measured quarterly flow and the adjustment factor. This adjustment was done for the six gages where the related intake was more than 1.6 km (1 mile) away, or where incoming tributaries contributed significant flow between the sites; adjustment factors ranged from 0.89 to 1.25. Average annual streamflow in the watershed was calculated based on the average streamflow at the PWS intakes.

Annual and quarterly *de facto* reuse proportions were calculated for each PWS based on the median annual streamflow and quarterly streamflow (median streamflow provided a more realistic estimate for the periods in this study). In this manner, earlier models (Barber *et al.* 2019) were used and adapted to report annual or quarterly ACCWW for the intake stream reaches based on measured streamflow. All derived estimates from this work, including adjustment factors, and annual and quarterly *de facto* reuse, are stored at the publicly accessible George Mason University Data Archive (Weisman 2021). Calculation of the proportion of accumulated wastewater in the watershed was based on the assumption that all wastewater entering a river segment was carried through to all downstream locations, which was consistent with that used by others previously (e.g., Rice & Westerhoff 2015). Wastewater quality was not addressed.

DBP compliance monitoring data (i.e., for THM4 and HAA5) from 2002 to 2019 were compiled for all PWSs in the study. The DBP data used came from locations within the distribution systems – i.e., where the DBP concentrations are most relevant to consumers. The THM4 and HAA5 concentrations used in the study were calculated on the basis of the average concentration at all monitoring locations for the quarter. The annual average for each PWS was calculated as the numerical average for the four quarters. Annual average DBP concentrations in the watershed were calculated from the average concentrations at the PWSs. Finally, as THM4 and HAA5 compliance monitoring data were available only for the period 2002–2019 for which WWTP flow data were available, the study was focused on this 18-year period of record (POR).

RESULTS AND DISCUSSION

The annual WWTP flow values over the POR (Figure 2(a)) show that flows were relatively constant for most of the period with a small upward trend toward the end. The flows ranged from 0.024 to 0.035 m^3/sec (0.54–0.81 MGD) – mean = 0.028 m^3/sec, median = 0.027 m^3/sec (mean = 0.63 MGD, median = 0.62 MGD). Both

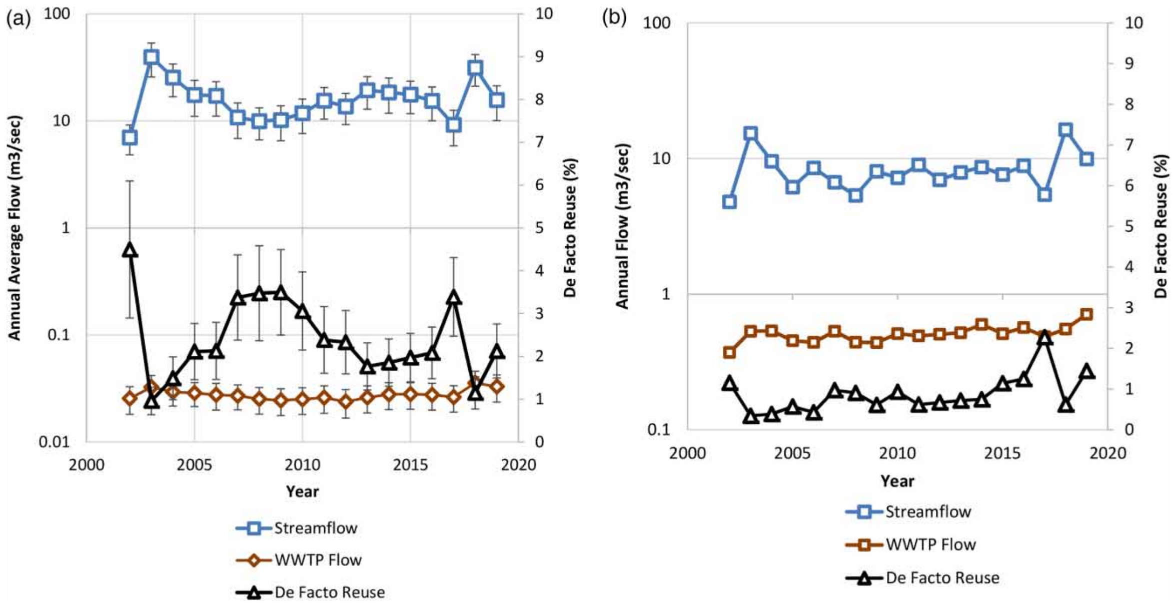

Figure 2 | Average annual (a) streamflow and *de facto* reuse among 11 PWSs and 98 WWTPs in the Shenandoah River watershed, with standard error, and (b) at a specific WWTP near USGS Gage #01622000.

streamflow and *de facto* reuse varied more than WWTP flow, and have the expected inverse relationship. Taken annually over the POR, streamflow ranged from 7.0 to 39.4 m^3/sec (160–900 MGD) – mean = 17.1 m^3/sec, median = 15.8 m^3/sec (mean = 390 MGD, median = 360 MGD) and *de facto* reuse ranged from 0.96 to 4.5% (mean = 2.4%, median = 2.1%). Streamflow was relatively higher in both the earlier and later parts of the POR, while *de facto* reuse was lower over those time periods. The Kruskal–Wallis test (a non-parametric method), showed that streamflow varied significantly from year to year over the POR ($p = 0.00087$), but WWTP flow and *de facto* reuse, examined separately, did not do so. Figure 2 also shows the variations (as standard error) in annual streamflow and *de facto* reuse among the PWSs (n = 11) and WWTP flow among the WWTPs (n = 98) in this study. Annual WWTP flow variations were examined for the WWTP with the highest annual average flow (Figure 2(b)). Like the other WWTPs, annual flow was influenced by the I&I associated with heavy rain events – for example, in 2018. Figure 2(b) also shows the annual streamflow variations at USGS Gage #01622000, (drainage area approximately 974 km^2 (376 square miles)) and variations in *de facto* reuse at a nearby PWS intake. The WWTP outfall is downstream of the PWS intake and thus not reflected in the *de facto* reuse at the PWS. As in Figure 2(a), these data also show streamflow and *de facto* reuse varying more than WWTP flow.

The average annual concentrations for both THM4 and HAA5 were relatively constant over the POR, apart from one relatively high THM4 value at the start (Figure 3). THM4 ranged from 36 to 62 micrograms per liter (μg/L) (mean = 43, median = 41) and HAA5 from 21 to 39 μg/L (mean = 33, median = 34). The Kruskal–Wallis test also showed that neither parameter varied significantly. Figure 3 also shows the standard error in annual average concentrations among the 11 PWSs. The average concentrations for both groups of chemical species were below the U.S. federal maximum contaminant levels (MCLs) of 80 (THM4) and 60 (HAA5) μg/L, respectively (USEPA 2010). As discussed previously, source water bromide, an important DBP precursor, was relatively low in the watershed, so no evaluation of bromide-based DBP species was conducted.

The range of streamflow, WWTP flow, and *de facto* reuse values varied on a quarterly basis throughout the POR. At quarterly level, streamflow ranged from 0.13 to 83 m^3/sec (3.0–1,900 MGD) – mean = 19.7 m^3/sec, median = 9.2 m^3/sec (mean = 450 MGD, median = 210 MGD) (Figure 4(a)), WWTP flow from 0.0014 to 530 liters per second (L/sec) (0.000031–12 MGD) – mean = 25 L/sec, median = 1.6 L/sec (mean = 0.56 MGD, median = 0.036 MGD) (Figure 4(b)), and *de facto* reuse from zero to 12% (mean = 2.8, median = 1.8) (Figure 4(c)). Values in Figure 4(a) and 4(b) that are higher than the 'whisker' are plotted individually.

In Quarter 3, median values for streamflow, WWTP flow and *de facto* reuse were 6.1 m^3/sec (140 MGD), 1.6 L/sec (0.037 MGD) and 4.3%, respectively. Separate Kruskal–Wallis tests on quarterly streamflow and *de facto* reuse, showed significant variation ($p = 1.0 \times 10^{-14}$ and 7.8×10^{-9}, respectively), while WWTP flow

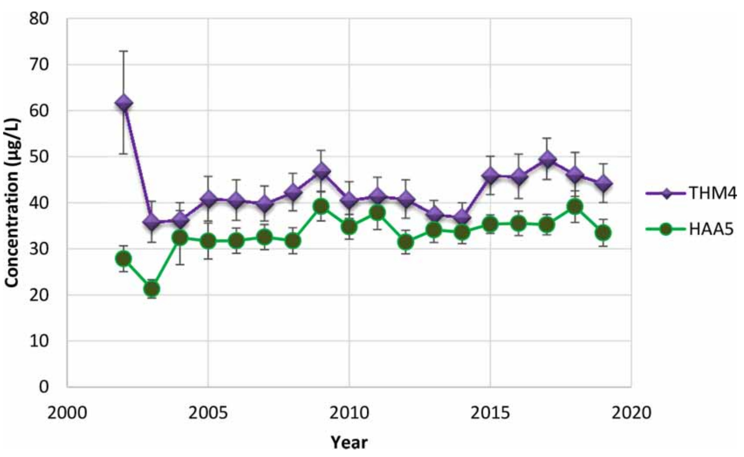

Figure 3 | Average annual DBP concentrations among 11 PWSs in the Shenandoah River watershed, with standard error.

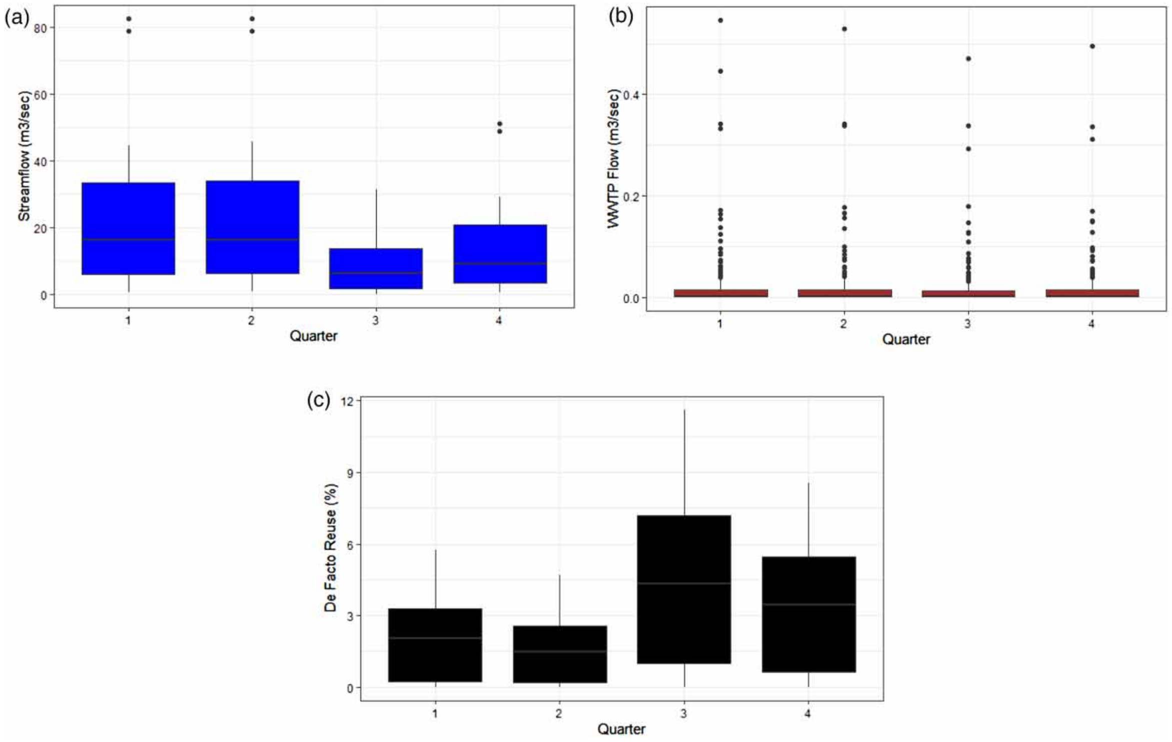

Figure 4 | Quarterly value ranges for (a) streamflow (n = 44), (b) WWTP flow (n = 376), and (c) *de facto* reuse (n = 44) at PWS intakes in the Shenandoah River watershed (2002–2019).

showed no significant variation. Dunn's test (also known as Bonferroni t) showed that there were significant quarterly differences for streamflow and *de facto* reuse. Streamflow in quarters 3 and 4 differed significantly from the other three quarters, while *de facto* reuse in quarters 3 and 4 each differed significantly from quarters 1 and 2, but quarter 3 did not differ significantly from quarter 4.

The THM4 and HAA5 median quarterly concentrations at PWS intakes, respectively, ranged from approximately 25 to 65 µg/L and 25 to 40 µg/L (Figure 5). HAA5 concentrations generally varied less by quarter than those for THM4. Separate Kruskal–Wallis tests on quarterly THM4 and HAA5 data showed that they vary significantly between quarters (for both, $p = 2.2 \times 10^{-16}$). Dunn's test for THM4 showed that quarters 1 and 3 differed significantly from all other quarters, while quarters 1 and 4 differed significantly from all other quarters for HAA5. THM4 concentrations in Quarter 2 that were lower than the 'whisker' are plotted individually.

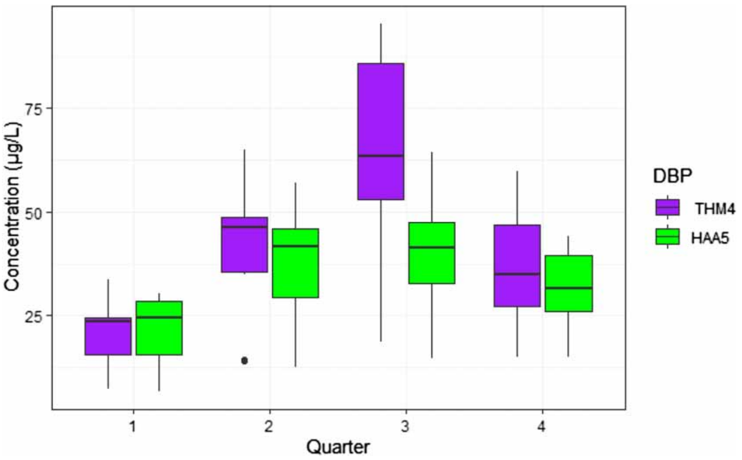

Figure 5 | Range of average quarterly concentrations for THM4 (n = 44) and HAA5 (n = 44) at Shenandoah watershed PWSs (2002–2019).

The concentrations of both THM4 and HAA5 increased as a function of *de facto* reuse on an annual average basis (Figure 6). THM4 concentrations, as a function of *de facto* reuse, were generally highest in quarter 3, and progressively lower in quarters 2, 4, and 1 (Figure 7(a)). The quarterly relationships for HAA5 differed from those for THM4 when *de facto* reuse exceeded 1%. Under that condition, HAA5 concentrations as a function of *de facto* reuse were generally highest in quarter 2, and then progressively lower in quarters 3, 4, and 1 (Figure 7(b)). The HAA5 concentration is affected more than that of THM4 by several factors, which may have led to the differences observed: (1) the hydrophobicity of natural organic matter (NOM) (greater hydrophobicity is associated with higher HAA5 concentrations (Solarik *et al.* 2000; Liang & Singer 2003); and (2) the relatively lower temperatures in quarter 2 than 3. HAA5 is susceptible to biodegradation, which would be likely to occur faster at the higher temperatures in quarter 3 (Bayless & Andrews 2008).

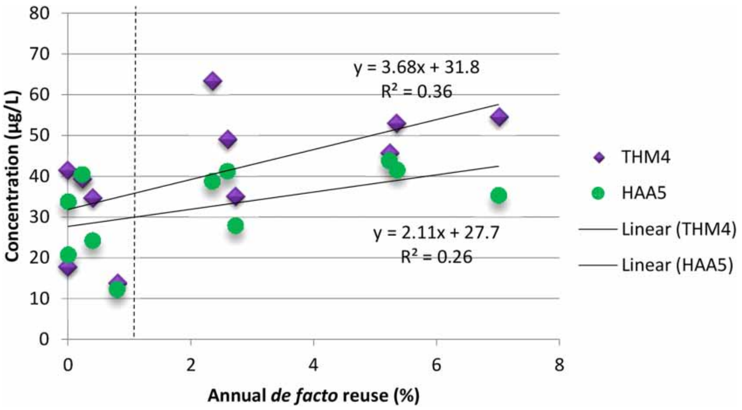

Figure 6 | Average annual DBP concentrations as a function of average annual *de facto* reuse. (Lines are linear models of annual averages; data points represent mean annual average values for the 11 PWSs (2002–2019)).

As shown in Table 1, THM4's linear relationship with *de facto* reuse was statistically significant for the annual average ($p = 0.050$, r = 0.54), and quarters 3 ($p = 0.032$, r = 0.59) and 4 ($p = 0.031$, r = 0.60), the quarters when *de facto* reuse was at its highest. The linear relationship of THM4 concentration annual average 75-percentiles with proportional *de facto* reuse (%) was also statistically significant for the annual average ($p = 0.038$, r = 0.57), and quarters 1 ($p = 0.035$, r = 0.58), 3 ($p = 0.029$, r = 0.60), and 4 ($p = 0.043$, r = 0.56). The linear relationship of HAA5 with *de facto* reuse was only significant statistically for quarter 1 at the 25th percentile ($p = 0.039$, r = 0.57), and nowhere else.

Using a t-test with average values for each PWS, significant differences in DBP levels for PWSs relative to 1% *de facto* reuse were examined. Table 2 shows that there was a significant difference ($p < 0.05$) in DBP

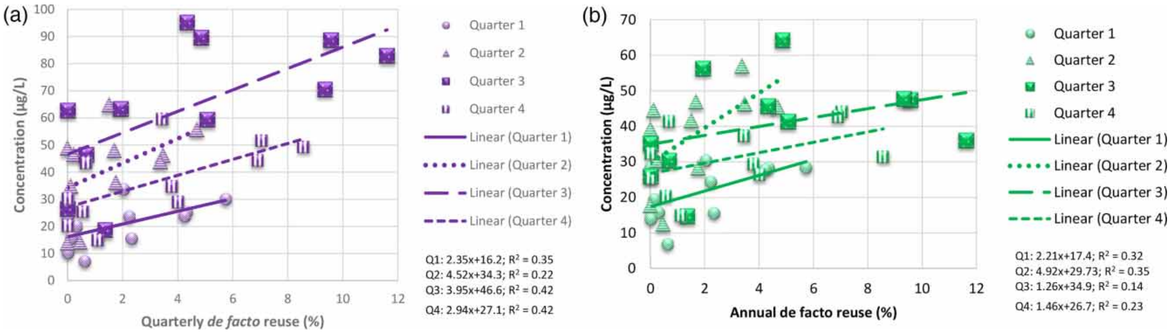

Figure 7 | Average quarterly concentrations for (a) THM4 and (b) HAA5 as a function of *de facto* reuse. (Lines are linear models of quarterly averages; data points represent mean quarterly average values for the 11 PWSs (2002–2019).)

Table 1 | Significance of linear relationship between DBP class and *de facto* reuse at the Shenandoah River watershed PWSs

DBP class	Annual average	Quarter 1	Quarter 2	Quarter 3	Quarter 4
Mean values					
THM4	**YES**	NO	NO	**YES**	**YES**
HAA5	NO	NO	NO	NO	NO
75th percentile					
THM4	**YES**	**YES**	NO	**YES**	**YES**
HAA5	NO	NO	NO	NO	NO
25th percentile					
THM4	NO	NO	NO	NO	NO
HAA5	NO	**YES**	NO	NO	NO

Table 2 | Significance of linear relationship between DBP class relative to 1% *de facto* reuse at Shenandoah watershed PWSs

DBP Class	Annual Average	Quarter 1	Quarter 2	Quarter 3	Quarter 4
Mean values					
THM4	**YES**	**YES**	NO	NO	NO
HAA5	NO	NO	NO	**YES**	NO
75th percentile					
THM4	**YES**	**YES**	NO	NO	NO
HAA5	NO	NO	NO	**YES**	NO
25th percentile					
THM4	**YES**	NO	NO	**YES**	NO
HAA5	NO	NO	NO	NO	NO

concentrations above or below this threshold for the annual average and quarter 1 THM4 concentrations, and for quarter 3 HAA5 concentrations, as well as at the 75-percentile level. At the 25-percentile level, the annual average and quarter 3 THM4 concentrations also showed a significant difference. This implies that a 1% *de facto* reuse value is an important threshold in annual average THM4 concentrations in PWSs in the watershed (i.e., with elevated annual average levels of DBP precursors) but that quarterly THM4 and HAA5 concentrations may be influenced more by factors such as temperature or pH than the proportion of reuse.

Temporal variations in WWTP flow, streamflow, *de facto* reuse, and DBPs, by year and quarter, are relevant to the water quality at PWSs in the watershed. Combined data from multiple state and federal sources have shown that a temporal evaluation of DBPs in PWSs can be related to the influence of *de facto* reuse from upstream WWTPs. The relationship of *de facto* reuse with DBPs found is generally consistent with previous studies

(Weisman *et al.* 2019). However, consideration of temporal variations in this study yielded some new findings. For example, both studies – this and Weisman *et al.* (2019) – showed that the annual average THM4 concentration had a statistically significant linear relationship with *de facto* reuse and that a *de facto* reuse level of at least 1% was associated with significantly higher concentrations of DBPs. Both studies showed that THM4 concentrations also had a statistically significant linear relationship with *de facto* reuse under low streamflow conditions (i.e., quarter 3), but that 1% *de facto* reuse was not associated with significantly higher THM4 concentrations under low streamflow conditions. In contrast to the 2019 study, 1% *de facto* reuse was not associated with significantly higher annual average HAA5 concentrations, except during quarter 3, even though the concentration of HAA5 varied less than that of THM4 across the POR.

The *de facto* reuse results from this study are consistent with those reported in Wiener *et al.* (2020) – an examination of temporal *de facto* reuse and streamflow variations by year and quarter for the Wabash River watershed (comprising parts of Illinois, Indiana, and Ohio). That study showed a wide range of water reuse (3–134%) with substantial quarterly variations (highest values in quarter 3). The major wastewater discharges into the Wabash watershed are from thermoelectric power plants (average 79% of all water discharged), a higher fraction of industrial discharges than found among the larger WWTPs in the Shenandoah watershed (none of the larger Shenandoah watershed WWTPs reported power plant discharges at a relative volume comparable to those for the Wabash watershed). Wiener *et al.* (2020) did not examine DBPs at PWSs in the Wabash River watershed.

The approach used to adjust streamgage flows to represent river conditions at PWS intakes before calculating the proportion of ACCWW is an innovative way to estimate *de facto* reuse using real-time streamflow fluctuations for this watershed. Prior research showed that streamflow varies substantially at the watershed gages – for example, at USGS Gage #01636500 (Shenandoah River), streamflow varied by a factor exceeding three (24.5–83.2 m^3/sec) (560–1,900 MGD) on an annual average basis and exceeding two (17.7–39.9 m^3/sec) (410–910 MGD) on a quarter 3 basis (Krstolic 2015), from the 25- to 75- percentiles. By relating ungaged PWS intakes to the nearest streamflow station, whether upstream or downstream, the use of historical streamflow gage data was maximized, improving accuracy.

Information about the *de facto* reuse and DBP formation relationship can also help to inform decision-making about wastewater treatment investments as a means of improving downstream water quality (Keiser & Shapiro 2019). Information about temporal *de facto* reuse variations can also be used by those seeking to control DBPs better, especially when there are concerns about elevated DBP levels.

Statistical evaluation of multiple data sets has been used to evaluate temporal and spatial variability of water quality (Singh *et al.* 2004; Summerhayes *et al.* 2011), but care is needed with secondary use of multi-source water quality data (Sprague *et al.* 2017). In this study, regulatory compliance data for WWTP flows and DBP concentrations were combined with high-quality streamgage data and a comprehensive watershed model, and provided a realistic mechanism for relating *de facto* reuse to DBPs, demonstrating potentially meaningful results for PWS operators.

Total organic carbon (TOC) is another indicator for DBP precursors in addition to *de facto* reuse. For conventional, surface water-supplied PWSs like those studied, U.S. federal drinking water regulations require that TOC

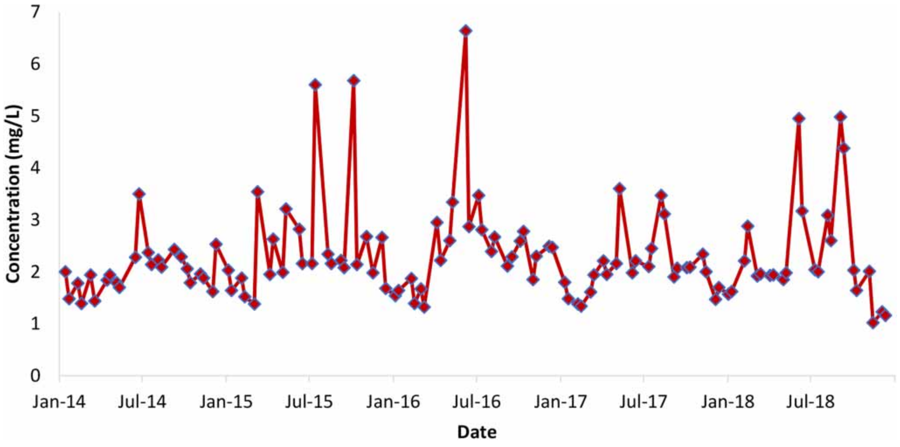

Figure 8 | TOC concentration variability at a PWS intake in the Shenandoah watershed from 2014 to 2018.

be monitored at the intake (USEPA 2010). TOC monitoring data were examined for the PWS included in this study, which had the highest proportional *de facto* reuse among those studied; TOC concentration data for their intake were compiled twice each month for 5 years (2014–2018) (Figure 8). Source water TOC concentrations ranged from approximately 1 to 7 mg/L (mean = 2.3; median = 2.1); the higher concentrations occurred in quarter 3 and are consistent with the types of precursor loads expected with the higher proportional *de facto* reuse estimated for that quarter.

CONCLUSIONS

The study showed annual and quarterly temporal variations in *de facto* reuse and DBPs at PWSs in the Shenandoah River watershed. The approach used to adjust gaged flow to represent river conditions on ungaged reaches and subsequently to estimate streamflow fluctuations used in calculating *de facto* reuse at PWS intakes is innovative. Maximizing the use of detailed historical USGS streamgage data enabled a more detailed streamflow evaluation and improved the final results.

Quarterly temporal variations were more pronounced than those between years over the 18-year POR. More extensive temporal variation analysis also supported the linear relationship between *de facto* reuse and THM4 concentrations, and the 1% *de facto* reuse threshold value.

This study of the Shenandoah River watershed was limited to conditions with relatively modest levels of *de facto* reuse and DBPs, compared to those in national studies (Rice & Westerhoff 2015; Seidel *et al.* 2017), thus making it more difficult to discern the potential effects of selected factors on the relationships between *de facto* reuse and DBPs.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the contributions from Stig Regli and Dr Adam Cadwallader at USEPA; Steffanie Keefe and Emily Majcher at USGS; and other reviewers who helped improve the paper's quality. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

FUNDING

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

DATA AVAILABILITY STATEMENT

All relevant data are available from an online repository or repositories at https://doi.org/10.13021/orc2020/YIQTTG.

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First received 23 May 2021; accepted in revised form 21 July 2021. Available online 4 August 2021