

# ARTICLE OPEN

# Drinking water vulnerability in less-populated communities in Texas to wastewater-derived contaminants

Thuy T. Nguyen<sup>1</sup> and Paul K. Westerhoff o

De facto potable reuse occurs when treated wastewater is discharged upstream of drinking water treatment plants (DWTPs) and can lead to contaminants of emerging concern (CECs) occurring in potable water. Our prior research, focusing on larger communities that each serve >10,000 people across the USA, indicates that elevated de facto reuse (DFR) occurs in Texas, and thus we added to our model DWTPs serving smaller communities to understand their vulnerability to CECs. Here, we show that two-thirds of all surface water intakes in Texas were impacted by DFR at levels exceeding 90% during even mild droughts, and under average streamflow DFR levels range between 1 and 20%. DWTPs serving lower population communities (<10,000 people) have higher DFR levels, and fewer than 2% of these communities have advanced technologies (e.g., ozone, activated carbon) at DWTPs to remove CECs. Efforts to improve water quality in these less populated communities are an important priority. The model approach and results can be used to identify prioritization for monitoring and treatment of CECs, including in underserved communities, which normally lack knowledge of their impacts from DFR occurring within their watersheds.

npj Clean Water (2019)2:19; https://doi.org/10.1038/s41545-019-0043-0

# INTRODUCTION

Wastewater discharges into the natural environment can deteriorate surface water. In the United States of America (USA), the Clean Water Act regulates municipal wastewater discharges to keep the nation's surface waters quality fishable and swimmable. The US Environmental Protection Agency (USEPA) National Pollutant Discharge Elimination System (NPDES) regulates point source discharge of wastewater to surface waters, but it rarely considers impacts on downstream drinking water treatment plants (DWTPs). Studies have detected contaminants of emerging concern (CECs), including pharmaceuticals, personal care products, and industrial chemicals, originating from wastewater in DWTPs downstream of wastewater treatment plants (WWTPs). 1-5 A previous study on the 50 very large WWTPs (between 15 and 660 million gallons per days (MGDs)) across the US reported 6000 MGD (263 m<sup>3</sup>/s) discharging to surface waters and measured 56 active pharmaceutical ingredients in effluent samples.<sup>6</sup> Additionally, some CECs lead to N-nitrosodimethylamine (NDMA) disinfection by-product formation in drinking waters after chlorination.<sup>7-9</sup> Furthermore, the public perception of CECs is unfavorable, despite evidence of their minimal human health risk because of the low exposure potentials in drinking water. 10-13

De facto reuse (DFR) occurs when a municipality withdraws water from a river or reservoir that includes treated wastewater discharged from upstream WWTPs. 14,15 The previously developed De Facto Reuse Incidence Nations Consumable Supply (DRINCS) model 16 analyzed treated municipal wastewater discharges from WWTPs and included combined sewer systems, although it does not consider combined sewer overflows or wet weather by-passes. The DRINCS model has been used and validated through field sampling in several case studies. 5,15,17,18 Our prior DRINCS study concluded that >50% of DWTPs in the US serving 10,000 or more

people with treated surface water have at least one WWTP discharge upstream of the drinking water intake. <sup>16</sup> While the frequency of DFR is high, its magnitude is relatively low under average streamflow condition. That previous DRINCS study, which considered only DWTPs serving 10,000 people or more, found among the highest DFR occurs in the Texas Gulf region (US Geological Survey (USGS) Hydrologic Region 12) with DFR occurring at 90% of the DWTP intakes. Other studies also indicate high levels of wastewater in surface waters in Texas. <sup>19,20</sup> Therefore, this paper focuses on the state of Texas (USA) and DWTPs that contrasts larger (>10,000 people) to smaller (<10,000 people) sized communities.

CECs undergo biogeochemical transformations (e.g., hydrolysis, oxidation, hydroxylation, conjugation, cleavage, de-alkylation, methylation, and demethylation) in surface waters, and the transformations are impacted by stream geometry and travel times.<sup>21–23</sup> Transformation products are often more polar, less bioaccumulative, and can be less toxic than parent compounds in the aqueous environment. 15,18,24 However, some derivative compounds can be more persistent and may have adverse human health effects.<sup>25</sup> CEC removal at DWTPs depends on raw water quality, chemical structure of target CECs, and specific unit processes in place. 26,27 Prior DRINCS modeling and nearly all field campaigns to quantify DFR has focused on DWTPs serving larger populations (e.g., >10,000 people), thereby potentially overlooking impacts from DFR on DWTPs serving smaller (or potentially underserved) communities. The value of the data science approach behind DRINCS can allow screening or prioritization for CEC monitoring or treatment, after inclusion of DWTPs serving smaller-sized communities (<10,000 people) are included in the

<sup>1</sup>Nanosystems Engineering Research Center for Nanotechnology-Enabled Water Treatment, School of Sustainable Engineering and the Built Environment, Arizona State University, Tempe, AZ 85287-5306, USA

Correspondence: Paul K. Westerhoff (p.westerhoff@asu.edu)

Received: 8 May 2019 Accepted: 12 August 2019 Published online: 01 October 2019



In this study, we expanded the DRINCS model from only 156 DWTP intakes serving 10,000 or more people to include all DWTPs in Texas (US) by locating, ground truthing, and adding an additional 244 DWTP intakes serving 10,000 or fewer people. 16 De facto wastewater reuse was modeled under average and variable streamflow conditions. DRINCS only includes treated and discharged wastewater effluent, but not contributions from stormwater discharges or non-point sources (e.g., septic systems, surface runoff). Using a Dijkstra's algorithm, <sup>28</sup> proximal distances between WWTP discharges and DWTP intakes, and their frequency distribution when multiple WWTPs were located upstream, were incorporated for the first time into DRINCS. Using information about the specific unit processes installed at each DWTP, we evaluated the capability of the DWTPs to remove CECs, should they be impacted by upstream WWTP discharges (i.e., DFR). This information was then used to discuss social equity issues and the need to increase CEC monitoring in less populated, often rural, communities.

#### **RESULTS AND DISCUSSION**

DFR occurrence and magnitude under mean annual streamflows Figure 1 shows the spatially distributed levels of DFR under mean streamflow conditions for all DWTPs with surface water intakes within Texas. Two-thirds of the DWTP intakes (422 out of 595) were impacted by the potential presence of wastewater, as defined up having at least one upstream wastewater discharge. This includes 222 intakes at 182 DWTPs that serve populations of ≤10,000 (Table 1). DWTP intakes impacted by at least one upstream WWTP discharge included intakes located on lakes and reservoirs (n = 225), streams or creeks (n = 108), or canals (n = 89). While the frequency of DFR is high (~67%), roughly 60% of impacted DWTP intakes have <5% DFR under mean annual streamflows; 5% DFR equates to 5% of the water at a DWTP intake potentially being of wastewater origin based upon Eq. 2. However, DFR was higher in southwestern Texas with most having >20% DFR under annual mean streamflows. Thirty-four surface water intakes by DWTPs supplied by Rio Grande in the southwestern of Texas (Strahler stream order = 8) have high DFR (>20%). There were 173 surface water intakes by 97 DWTPs in Texas not impacted by upstream WWTP discharges, and 61 of these serve 10,000 or fewer people.

Strahler stream orders play an important role in DFR magnitude at drinking water sources. 16 Figure 2 shows that the highest DFR levels were in the smallest and largest rivers in Texas, or lower to higher Strahler stream orders. DFR varies substantially among DWTPs located on different stream orders. First-order streams are smaller and therefore rely more on WWTP discharges to maintain even mean annual streamflows. Hence, smaller streams are more likely to contain CECs throughout the year. Most DWTPs on second- through fifth-order streams had DFR below 5%. In contrast to national trends where DWTP intakes on higher stream orders have lower DFR,<sup>29,30</sup> presumably due to natural runoff diluting wastewater, streams of sixth, seventh, and eighth order in Texas show higher DFR. This illustrates how geographical location within the arid southwestern US can impact DFR perhaps more than general stream order classification on a national basis. Nearly all DWTPs on the higher stream orders are in Texas were impacted by at least one upstream wastewater discharge.

# Effects of variable streamflow on DFR magnitude

Reduced streamflow during drought may increase DFR. However, unlike the mandated requirement to have streamflow data or predictions at the WWTP discharge locations to calculate dilution factors, there are rarely in-stream gauging stations or long-term streamflow datasets available at DWTP intake locations. Lack of long-term (>30 years) data limits the ability to perform statistical

analysis of drought or flood impacts on DFR. With the use of USGS stream gauge database within National Hydrography Dataset (NHD) Plus suite, 22 of the 595 DWTP intakes had adequate long-term (>30 years) historical streamflow data, and DFR trends as a function of increasing streamflow were assessed. Figure 3 illustrates DFR for DWTPs as a function of both Strahler stream order and historical streamflow. Fifteen of the 25 sites have >10% DRF at the 50th percentile flow. At the 7Q2 condition (~10th percentile streamflow), treated wastewater made up ~100% of the water supply for 14 of 25 DWTP intakes. During seasonal low flow or drought periods, which is the design condition for WWTP effluents, there is a high occurrence of DFR (and associated CECs) at downstream drinking water intakes.

Proximity distribution of WWTPs upstream to DWTPs within the Trinity River Basin

Figure 4 shows the location of 151 WWTPs discharges upstream of 10 surface water DWTP intakes in the Trinity River Basin. Located at the upstream end of the lake, DWTP 10 withdrew water from Lake Lewisville, while DWTP 04 and DWTP 05 also used Lake Livingston as drinking water source. Other DWTPs withdraw water from tributaries (Elm Fork Trinity River) or mainstream of the Trinity River.

Twenty-one WWTPs influenced the most up-river drinking water facility (DWTP 10), whereas 151 WWTP discharges were upstream of DWTP 01. Because WWTPs discharge into tributaries and the main stream of the Trinity River, linear addition of treated wastewater does not occur. Instead, there is a distribution of distances from different tributaries that affect an individual DWTP. Figure 5 and Table SI.3 present cumulative distributions for the number of WWTPs located at different distances upstream from each of the 10 DWTPs. Figure SI.4 and Table SI.2 show cumulative wastewater discharges, instead of number of facilities, using a similar x-axis. There are no DWTPs within this watershed with a WWTP located fewer than 16 km upstream (10 miles), except two facilities (DWTP 04 and 10) are located on lakes that receive WWTP discharge. Lakes can have complex stratification and mixing patterns and would necessitate site-specific hydrologic modeling to understand precise levels of DFR. However, DRINCS helps identify such site-specific needs. Eight DWTPs have WWTPs located 16-40 km upstream (10-25 miles), and most of the WWTP discharges are located 160-500 km (100-300 miles) upstream of DWTP intakes. Figure SI.4 illustrates cumulative wastewater upstream for each of ten DWTPs in Trinity River basin. The wastewater volume varied from <10 MGD (within <16 km) to nearly 1400 MGD (>1600 km).

Travel time of CECs in rivers can reduce their concentrations through biogeochemical transformations. Travel time can be calculated by dividing distance by streamflow velocity. However, velocities depend on volumetric flowrate, drainage area, rainfall intensity-frequency-duration relationships, gradient or slope of the riverbed, and cross-sectional area of the channel. For lakes and reservoirs, NHD Plus identified streamlines were used to calculate travel times and then CEC attenuation; more detailed lake mixing models could be pursued in the future that include lake stratification or mixing and hydraulic residence times. High velocities often occur during flood or other high streamflow events, where greater wastewater dilution occurs and thus is probably less important for CECs than lower flow periods.<sup>2</sup> Typical stream velocities are 0.15-0.6 m/s (0.5-2 ft/s), but they can be slower under low streamflow conditions. Travel time estimates are shown in Fig. Sl.5. A streamflow of 0.3 m/s would result in travel times of 0.6, 1.5, 6.2, and 19 days for 16, 40, 160, and 500 km, respectively. CEC half-lives in surface waters can range from hours (e.g., photo-labile) to months (e.g., artificial sweeteners), depending upon their reactivity. For seven CECs commonly used as surrogates,<sup>31</sup> we applied EPI Suite<sup>TM</sup> and fate model LEV3EPI<sup>TM</sup> to

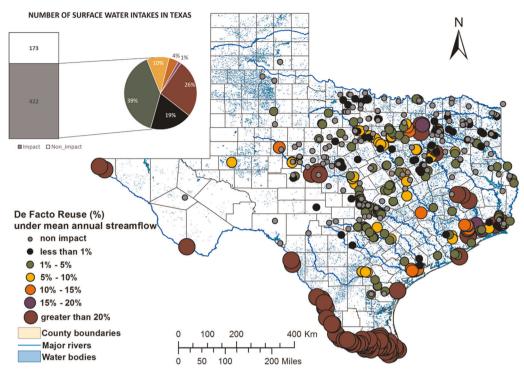


Fig. 1 Drinking water treatment plants (DWTPs) affected by upstream wastewater treatment plants (WWTPs) discharge under mean annual stream flows in Texas

Description	Values categorized by USEPA DWTP sizes					Totals
	Very small	Small	Medium	Large	Very large	
Population served	≤500	501–3300	3301–10,000	10,001–100,000	>100,000	~19 Million
Surface water facilities						
# Intakes (# impacted <sup>a</sup> )	60 (39)	148 (99)	117 (84)	192 (133)	78 (67)	595 (422)
# DWTPs (# impacted <sup>a</sup> )	49 (34)	114 (86)	82 (62)	127 (96)	28 (25)	400 (303)
DWTP with advanced Tech <sup>b</sup> in	n category size					
From All DWTPs	0%	2.6%	4.9%	7.1%	18%	5.3%
Only impacted DWTPs <sup>a</sup>	0%	3.5%	6.5%	7.3%	20%	4.8%

DWTPs drinking water treatment plants, USEPA US Environmental Protection Agency

estimate half-lives.<sup>32</sup> Table SI.4 shows the degradation rates of the seven CECs in water. Of the compounds studied, ibuprofen had the shortest half-life in water (15 days); diclofenac, meprobamate, gemfibrozil, and sulfamethoxazole SMX were next at 37.5 days; and tris(1-chloro-2-propyl) (TCPP) and tris(2-chloroethyl) (TCEP) had the longest half-life (60 days). CEC attenuation with distance was estimated using pseudo-first-order degradation kinetics:<sup>33</sup>

$$C(t) = C_i * e^{-kt}, \tag{1}$$

where C(t) is the analyte concentration at time t,  $C_i$  is the initial analyte concentration, k is the first-order transformation rate (1/day) and  $k = \frac{\ln 2}{t_{1/2}}$ ,  $t_{1/2}$  is the half-life of CEC in water (days), and t the travel time (days), calculated as distance divided by streamflow velocity.

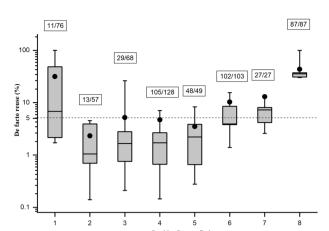
Typical streamflow velocities range between 0.05 and 0.5 m/s, resulting in travel times of 6–60 days for a proximal distance of

250 km. Figure SI.6 shows as a function of distance the degradation of several CECs commonly used as WWTP surrogates (meprobamate, ibuprofen, gemfibrozil, diclofenac, sulfamethoxazole, TCPP and TCEP phosphates) for larger CEC lists that may number in the hundreds of compounds. <sup>5,17,31,34</sup> For a 0.1 m/s streamflow, roughly 50% of the ibuprofen degraded within 100 km, whereas 50% degradation of diclofenac, meprobamate, gemfibrozil, or sulfamethoxazole may not be reached until 300 km. Even longer distances (600 km) may be required for similar degradation of TCPP or TCEP.

Streamflow variation impacts levels of CECs at downstream DWTP intakes in two ways: (1) lower streamflow proportionately increases CEC concentrations in rivers just below WWTP discharges (i.e., less dilution), but (2) lower streamflows proportionately lengthens hydraulic travel times that allow for more CEC attenuation via biogeochemical transformations. For the ten

<sup>&</sup>lt;sup>a</sup>Indicated values are for facilities impacted by de facto reuse

<sup>&</sup>lt;sup>b</sup>Advanced technology is defined as using ozonation or with hydrogen peroxide granular activated carbon, or reverse osmosis



**Fig. 2** De facto reuse (DFR) magnitude at drinking water treatment plant (DWTP) intakes under average flow condition in Texas (top and bottom of box = 75th and 25th percentiles, respectively; top and bottom of whisker = 90th and 10th percentiles, respectively; line inside box = 50th percentile (median); dot (•) = average; dashed line = 5% DFR). Numbers above each bar-and-whisker diagram indicate the number of DWTP intakes with DFR >0 included in the analysis for each stream order relative to the total number of DWTPs in Texas on surface water supplies having that stream order

DWTP intakes considered in Fig. 5 where CEC transformations similar to that predicted over 300 km may occur (Figs. Sl.5 and Sl.6), nine DWTPs had between 20 to 30 upstream WWTP dischargers within 161 to 483 km and DWTP#7 had 65 WWTP discharges within that distance. With the distances between DWTPs downstream from multiple WWTP discharges ranging from <16 to >800 km (Fig. 5), it is probable that some CECs will be largely removed by natural attenuation, while concentration of more refractory CECs (e.g., TCEP, TCPP) would likely be relatively unchanged at the downstream DWTP intake.

# Unit processes at DWTPs impacted by de facto reuse

Water treatment plants can build and operate advanced unit processes capable of removing CECs from the intake water, in addition to conventional unit processes required to meet existing regulatory compliance. However, CECs are by their very nature "emerging" and not currently regulated. Therefore, few DWTPs are required to install advanced unit processes, unless for secondary benefits (e.g., reduction in algae-derived tastes and odors) or necessity to meet disinfection and disinfection by-product rules established by the USEPA. This section uses data from the State of Texas on the type of unit processes installed at DWTPs to explore which facilities, as a function of their size and impact by DFR, employ advanced unit processes, which would be able to remove CECs. Figure 6 and Table SI.5 summarize the unit processes installed at all DWTPs in Texas and also for the subset of DWTPs impacted by DFR. Each DWTP combines several unit processes that will achieve variable CECs removal efficiencies. Two hundred and thirty-six DWTPs impacted by wastewater in Texas disinfect using chloramines. DWTPs using free chlorine can also form chloramines if ammonia from upstream WWTPs is present. Chloramines react with some CECs to produce NDMA and other probable carcinogens.<sup>7</sup> Prior work shows correlations between detectable NDMA at DWTPs with DFR >0,8 suggesting that CEC removal may be necessary. State-of-the-art unit process trains for planned, direct potable reuse include (1) reverse osmosis followed by advanced oxidation; (2) river bank filtration; or (3) ozonation followed by biofiltration, followed by an environmental (groundwater aquifer, surface water) or engineered buffer. 35-37 However, comparable strategies currently do not exist for DWTPs with DFR.

Conventional treatment processes (i.e., coagulation, sedimentation, and filtration) are used at >80% of the DWTPs in Texas. However, the conventional unit processes achieve <30% CEC removal.<sup>26</sup> Ultra- or microfiltration provide only minimal improved performance in CEC removal compared against granular media filtration. Advanced oxidation processes (e.g., ozonation or ultraviolet (UV) irradiation alone or with hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>)) are effective in removing CECs.<sup>26,31,38</sup> However, only 13 of 303 (5%) DWTPs that are impacted by DFR use these unit processes; DWTPs use ozonation alone (n = 10) or with hydrogen peroxide (n = 2), and UV with hydrogen peroxide (n = 1). Physical removal of CECs can be achieved by sorption to activated carbon or separation using nanofiltration or reverse osmosis membranes. 39-43 Forty-three of 303 DWTPs in Texas with DFR >0 use activated carbon (both in granular and powder one) and only nine of those DWTPs impacted by DFR use granular activated carbon (GAC). GAC is often used at DWTPs to control algal-related taste and odors, DBP precursors, and more recently CECs. Seven DWTPs impacted by DFR report using of reverse osmosis.

# DWTP treatment disparity for low population communities impacted by DFR

Many DWTPs (N = 303) in Texas are impacted by at least one upstream WWTP (Fig. 1), including 182 DWTPs serving 10,000 or fewer people, of those are 120 DWTPs serving 3300 or fewer people. However, because the advanced DWTP unit processes are not uniformly applied at smaller and larger DWTPs, CEC exposure in treated drinking waters varies. Figure 7 shows the distribution of DWTP levels of treatment by population served, and whether the DWTP is impacted or not by DFR. Figure 7 also superimposes whether or not the unit processes at the DWTP are capable of removing CECs (i.e., advanced treatment). For this analysis, we considered advanced treatment processes those with the highest potential to remove CECs: ozone alone or with hydrogen peroxide, GAC, or reverse osmosis. As summarized in Table 1, the majority of DWTPs serving smaller communities (<10,000 people) did not employ advanced treatment (Fig. 7), and the percentage not employing advanced treatment was even higher (90%) among the smallest DWTPs (serving <3300 people). Less populous communities with smaller DWTPs often lack the financial capacity (e.g., taxation base) to fund capital investment and higher operational costs associated with advanced treatment unit processes. Needs clearly exist to provide financial mechanisms to encourage installation of more advanced drinking water processes at "higher-risk" DWTPs (i.e., those with higher DFR).

There are many reasons advanced technologies are not installed at facilities serving smaller communities (<3300 or 3300-10,000). The disparity in drinking water quality in systems serving smaller versus larger populations is evident in the number of violations across the US for existing USEPA regulations.<sup>44</sup> For example in Texas, Table SI.7 and SI.8 shows nearly 70% of the maximum contaminant level violations occur at systems serving fewer than 10,000 people. Figures SI.9 and SI.10 show that the most commonly reported violations are total trihalomethanes and five haloacetic acids (HAA5). By their very nature CECs are "emerging" and hence are unregulated; thus, they are not part of health-based water quality violations at DWTPs. 45 We considered relationships between cancer mortalities and DFR, but as many CECs are pharmaceuticals they do not cause cancer, but rather potentially endocrine disruption or a number of other endpoints that have yet to be epidemiologically supported at low concentrations that occur in drinking waters.<sup>12</sup> One use of this paper could be to locate potential communities for inclusion in such toxicology<sup>17,46</sup> or epidemiology studies.

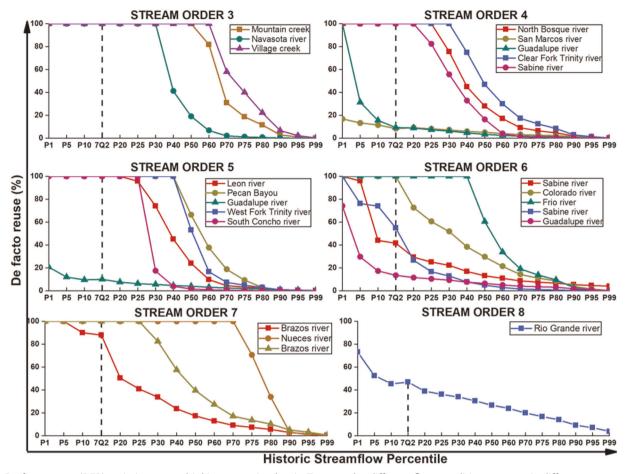


Fig. 3 De facto reuse (DFR) variation at 22 drinking water intakes in Texas under different flow conditions across six different stream orders. Exact drinking water treatment plant (DWTP) locations on each stream are not shown to protect the utility confidentiality

# **Implications**

This study found that 303 DWTPs in Texas were impacted by at least one upstream WWTP (Fig. 1), including 182 DWTPs serving <10,000 people with 120 of those DWTPs serving <3300 people. Smaller communities are more commonly located on lower stream order (Strahler stream order first to fifth; Fig. Sl.7). Thus, more of small DWTPs are likely impacted by CECs in Texas. Using similar methodologies as applied herein, DFR levels covering the same orders of magnitude as reported herein are being predicted globally. However, those studies did not focus on impacts to smaller utilities, travel times between WWTP discharges and DWTP intakes, or relate the type of treatment to the presence of CECs in DWTP intake or treated waters.

Because CECs can be transformed within surface waters, we analyzed the frequency distribution of upstream proximal distance of WWTPs discharge locations from downstream DWTP intakes. The Trinity River basin in Texas has 151 WWTPs and 45 DWTPs, and was used as further modeled to understand proximity of DWTPs from WWTPs. Most WWTPs were located 160 to 500 km upstream of DWTP intakes, where travel times between potential CEC sources and DWTP intakes range from 5 to 15 days under average streamflow. This leads to environmental exposures, but allows time for in-stream biogeochemical processes to transform some CECs.

This study also found that fewer than 10% of smaller sized DWTPs in Texas employ advanced technologies capable of removing CECs. Because these small communities have among the highest DFR levels, there is a need to increase resources to prioritize monitoring and installing advanced treatment in these

facilities. To date, most CEC field occurrence studies at DWTPs have involved only larger-sized facilities. There is a need to involve smaller-sized DWTPs in CEC occurrence studies to understand if small systems are disproportionately impacted by DFR. Analysis using DRINCS could help identify DWTPs at higher risk of DFR where such studies could be most beneficial in defining the magnitude of CEC occurrence. These may also be locations where investment in public infrastructure (e.g., upgraded WWTP or advanced DWTP unit processes) may have the largest ecological or human health risk, respectively. Until such infrastructure is installed, communities predicted to have high DRF levels may be of interest to the health community as locations for assessing biomarkers or health outcomes from wastewater reuse.

# **METHODS**

# Study area and facilities considered

Texas is located in the south-central USA, covers 695,662 km², and spans three national hydrologic units (Regions 11 (Arkansas White Red), 12 (Texas Gulf) and 13 (Rio Grande). Texas is the second-most populated state in the US, having about 25 million inhabitants in 2010. By 2060, the population is projected to double to 46 million people, and Texas's annual municipal water demand is predicted to increase from 4.9 million acre feet in 2010 to 7.8 million acre feet by 2060. Water availability varies in Texas, spanning from limited resources in the arid western region to being water-rich in eastern areas. Increasing populations will likely lead to greater reliance and impacts of planned and unplanned (de facto) wastewater reuse.

This study included 400 community public water systems in Texas withdrawing surface water from 595 surface water intakes (Table 1). Some DWTPs have more than one surface water intake. Figures SI.1 and SI.2 show locations of drinking water sources in Texas and the population served by



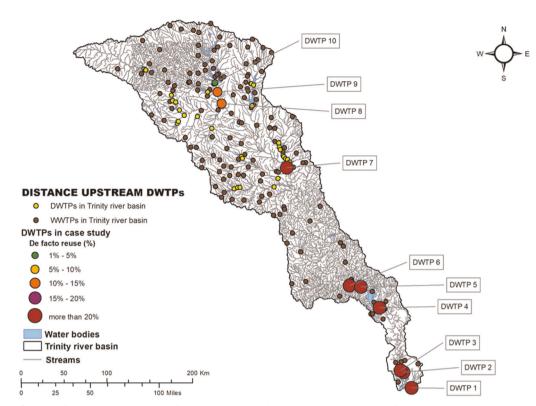
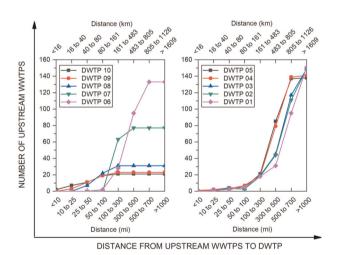


Fig. 4 Locations of ten drinking water treatment plants (DWTPs) used as a case study impacted by 151 upstream wastewater treatment plants (WWTPs) in Trinity River basin



**Fig. 5** Proximity analysis of ten drinking water treatment plants (DWTPs) using surface water in Trinity River basin (in terms of number of wastewater treatment plants (WWTPs) upstream)

each DWTP. These surface water DWTPs account for 2690 MGD of design capacity (118 m³/s). This is augmented with a large number of groundwater-supplied facilities that combine to treat up to another 1514 MGD (66 m³/s) of potable water. The groundwater facilities were not included in this study because DFR is less common in groundwater systems and is not considered in the DRINCS model. A DWTP database was retrieved, and activity codes for facilities unified, from the Texas Drinking Water Watch (TDWW), Texas Water Development Board (TWDB), and Texas Commission on Environmental Quality. The databases include DWTP intake locations (latitude and longitude), public water system identification number (PWSID), population served, and additional data. Information on type of installed treatment processes at DWTPs was obtained from the TDWW for the most recent 2-year dataset available (2017). The PWSID also

allows access to information on the unit processes at each facility. ArcGIS<sup>TM</sup> version 10.4 was used to create maps and conduct the spatial analysis.

The data that support the findings of this study were aggregated from a variety of US Federal sources. WWTP data were obtained from Clean Watersheds Needs Surveys 2008-Environmental Protection Agency, which includes facility name, permit number (NPDES), level of treatment, design capacity, and location (longitude and latitude of wastewater outfalls to surface water). There are 1206 WWTPs with a total design capacity of 3213 MGD (141 m³/s) that discharge to surface waters; an additional 253 facilities discharge to groundwater, ocean, or evaporation ponds. Figure Sl.3 shows that ~70% of the 1206 WWTPs included in DRINCS for Texas are relatively small, with treatment capacity below 1 MGD (0.05 m³/s).

# Predicting DFR using DRINCS

The ArcGIS-based model of DRINCS previously developed for all WWTPs and DWTPs serving 10,000 people or more 16 was augmented to include DWTPs serving 10,000 or fewer people from surface water sources. Precise locations of WWTP discharges and DWTP intakes were verified using the Texas Irrigation District Engineering and Assistance Program and visually ground-truthed using Google Earth. Streamflow data was obtained from the NHD-USGS, and stream networks were based on the mediumresolution NHD (1:100,000 scale). Strahler stream order defines stream size based on hierarchy of tributaries, 56,57 with values for the USA ranging from a low of first order to larger river networks that approach ninth order. Each river segment within a watershed is treated as a node, with the next segment downstream as its parent. For example, when two first-order streams join then a second-order stream forms. Strahler stream order can be obtained from additional calculated attributes in NHD Plus.  $^{56,58}$  DRINCS was also updated by adding USGS stream gauges from within the NHD Plus suite; attribute data include average, min, max, and percentile streamflows. A key objective was to maximize the available hydrologic datasets to cover the large possible variations based upon historic streamflows data. The statistical values were calculated based on the basis of the entire record period until 20 April 2004 (the date NHD pulled the data for analysis); the starting date for each gauging station varied depending upon when it began reporting data, with the earliest being 1 November 1915 and the latest was on 27 September 1997.

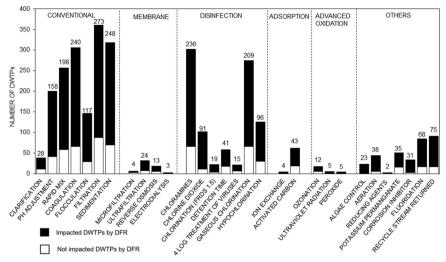
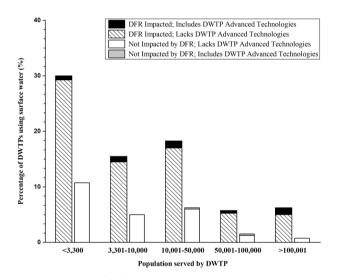


Fig. 6 Unit processes of drinking water treatment plants (DWTPs) using surface water in Texas (the number above each bar represents the number of DWTPs that are impacted by de facto reuse (DFR) and which implement that specific type of unit process)



**Fig. 7** Percentage of surface water drinking water treatment plants (DWTPs) in Texas categorized by population served. The percentage of non-impacted or impacted DWTPs using advanced technology was calculated as the number of DWTPs in each of four categories divided the total number of DWTPs in Texas. Advanced technology is defined as using ozonation or with hydrogen peroxide granular activated carbon, or reverse osmosis

DFR at each DWTP withdrawing surface water was calculated as the percent of treated wastewater at a particular surface water intake, following our previous published methods and assumptions:<sup>8,15,16</sup>

$$DFR = \frac{\sum WW}{Q} \times 100\%,$$
 (2)

where Q is the streamflow at the DWTP intake location, and WW is the accumulated discharge from all upstream WWTPs, calculated by running the Python script.

In Texas, the key discharge–frequency characteristic used to evaluate critical condition of the stream for ecological considerations is the "annual lowest mean discharge for seven consecutive days with a 2-year recurrence interval (7Q2)." <sup>59</sup> The 7Q2 low-flow index was calculated using an Excel-based application "calculator for low flow (CALF)," which was developed by Environmental Flows Information System for Texas based on daily stream flow for 30 years of continuous USGS gauging data via Hydrologic Information System; the default period to retrieve data with CALF is from 1 January 1940 through 31 December 2009. Values for 7Q2

must be reported at WWTP discharges, but are not required at DWTP intakes. Values of 7Q2 closely matched 10th percentile streamflows (from the calculations), and hence we considered low flow conditions as 10th percentile streamflows (Table SI.1—Supporting Information).

The proximity distribution between DWTPs and upstream WWTPs discharge locations was determined using digital stream networks with flow direction from the NHD Plus to build a geometric network for tracing upstream in ArcGIS. All the shapefiles were re-projected in a Texas-specific projection coordinate system in ArcGIS, namely Texas Centric Mapping System/Albers Equal Area. Vector datasets of upstream segments were then used to create ArcGIS Network Analyst tool that calculated the stream distance between WWTPs upstream from each DWTP. Once the network was constructed, the New Closest Facility analysis solver was able to identify the shortest routes along stream networks for each Facility (i.e., a single surface water DWTP intake) and Incidents (i.e., all upstream WWTP discharges) using Dijkstra's algorithm.<sup>28</sup> In this study, proximal distances were computed for 10 DWTPs, of which surface water intakes spanning along Trinity River in the case study in the Trinity River basin. The Trinity River was selected in part because it is one of the most populous watershed in Texas with a total area of 17,913 square miles for the 423 mile Trinity River (TWDB) and can contain >90% wastewater effluent under low flow conditions.60

#### **DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request. Sources of electronic dataset used within the paper are summarized in Table SI.9.

# **ACKNOWLEDGEMENTS**

This research was partially supported by the National Science Foundation (EEC-1449500, SES-1462086, CBET-1804229, and DEB-1637590), the Vietnam Education Foundation (VEF), Decision Center for a Desert City (DCDC), and ASU's Future H2O initiative. We would like to thank Julianna Acero for her assistance in ground-truthing DWTP and WWTP locations and Laurel Passantino for technical editing.

## **AUTHOR CONTRIBUTIONS**

T.T.N. conducted ground truthing and modeling, and wrote the majority of the paper. P.K.W. developed the idea and oversaw writing.

# **ADDITIONAL INFORMATION**

**Supplementary Information** accompanies the paper on the *npj Clean Water* website (https://doi.org/10.1038/s41545-019-0043-0).

Competing interests: The authors declare no competing interests.



**Publisher's note:** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

#### REFERENCES

- Benotti, M. J., Stanford, B. D. & Snyder, S. A. Impact of drought on wastewater contaminants in an urban water supply. J. Environ. Qual. 39, 1196–1200 (2010).
- Benotti, M. J. et al. Pharmaceuticals and endocrine disrupting compounds in US drinking water. Environ. Sci. Technol. 43, 597–603 (2009).
- Schultz, M. M. et al. Antidepressant pharmaceuticals in two US effluent-impacted streams: occurrence and fate in water and sediment, and selective uptake in fish neural tissue. *Environ. Sci. Technol.* 44, 1918–1925 (2010).
- Bieber, S., Snyder, S. A., Dagnino, S., Rauch-Williams, T. & Drewes, J. E. Management strategies for trace organic chemicals in water—a review of international approaches. *Chemosphere* 195, 410–426 (2018).
- Nguyen, T. et al. Modeled de facto reuse and contaminants of emerging concern in drinking water source waters. J. Am. Water Works Assoc. 110, E2–E18 (2018).
- Kostich, M. S., Batt, A. L. & Lazorchak, J. M. Concentrations of prioritized pharmaceuticals in effluents from 50 large wastewater treatment plants in the US and implications for risk estimation. *Environ. Pollut.* 184, 354–359 (2014).
- Hanigan, D. et al. Methadone contributes to N-nitrosodimethylamine formation in surface waters and wastewaters during chloramination. Environ. Sci. Technol. Lett. 2, 151–157 (2015).
- 8. Rice, J., Via, S. & Westerhoff, P. Extent and impacts of unplanned wastewater reuse in U.S. rivers. *J. Am. Water Works Assoc.* **107**, E571–E581 (2015).
- Rule, K. L., Ebbett, V. R. & Vikesland, P. J. Formation of chloroform and chlorinated organics by free-chlorine-mediated oxidation of triclosan. *Environ. Sci. Technol.* 39, 3176–3185 (2005).
- Rice, J., Wutich, A., White, D. D. & Westerhoff, P. Comparing actual de facto wastewater reuse and its public acceptability: a three city case study. Sust. Cities Soc. 27, 467–474 (2016).
- Anumol, T., Clarke, B. O., Merel, S. & Snyder, S. A. Point-of-use devices for attenuation of trace organic compounds in water. J. Am. Water Works Assoc. 107, E474–E485 (2015).
- Bruce, G. M., Pleus, R. C. & Snyder, S. A. Toxicological relevance of pharmaceuticals in drinking water. *Environ. Sci. Technol.* 44, 5619–5626 (2010).
- Stanford, B. D., Snyder, S. A., Trenholm, R. A., Holady, J. C. & Vanderford, B. J. Estrogenic activity of US drinking waters: a relative exposure comparison. J. Am. Water Works Assoc. 102, 55–65 (2010).
- 14. NRC. Water Reuse: Potential for Expanding the Nation's Water Supply Through Reuse of Municipal Wastewater 262 (National Research Council, Committee on the Assessment of Water Reuse as an Approach to Meeting Future Water Supply Needs, Washington, DC, 2012).
- Rice, J., Wutich, A. & Westerhoff, P. Assessment of de facto wastewater reuse across the US: trends between 1980 and 2008. Environ. Sci. Technol. 47, 11099–11105 (2013).
- Rice, J. & Westerhoff, P. Spatial and temporal variation in de facto wastewater reuse in drinking water systems across the USA. *Environ. Sci. Technol.* 49, 982–989 (2015)
- Barber, L. B. et al. Integrated assessment of wastewater reuse, exposure risk, and fish endocrine disruption in the Shenandoah River Watershed. *Environ. Sci. Technol.* 53, 3429–3440 (2019).
- Rice, J., Via, S. H. & Westerhoff, P. Extent and impacts of unplanned wastewater reuse in US rivers. J. Am. Water Works Assoc. 107, 93–93 (2015).
- Brooks, B. W., Riley, T. M. & Taylor, R. D. Water quality of effluent-dominated ecosystems: ecotoxicological, hydrological, and management considerations. *Hydrobiologia* 556, 365–379 (2006).
- Slye, J. L. et al. Relationships between benthic macroinvertebrate community structure and geospatial habitat, in-stream water chemistry, and surfactants in effluent-dominated Trinity River, Texas, USA. Environ. Toxicol. Chem. 30, 1127–1138 (2011).
- Challis, J. K., Hanson, M. L., Friesen, K. J. & Wong, C. S. A critical assessment of the photodegradation of pharmaceuticals in aquatic environments: defining our current understanding and identifying knowledge gaps. *Environ. Sci. Process. Impacts* 16, 672–696 (2014).
- Radke, M., Ulrich, H., Wurm, C. & Kunkel, U. Dynamics and attenuation of acidic pharmaceuticals along a river stretch. *Environ. Sci. Technol.* 44, 2968–2974 (2010).
- Chen, B. Y., Nam, S. N., Westerhoff, P. K., Krasner, S. W. & Amy, G. Fate of effluent organic matter and DBP precursors in an effluent-dominated river: a case study of wastewater impact on downstream water quality. Water Res. 43, 1755–1765 (2009).
- Boxall, A. B. A., Sinclair, C. J., Fenner, K., Kolpin, D. & Maud, S. J. When synthetic chemicals degrade in the environment. *Environ. Sci. Technol.* 38, 368A–375A (2004).

- Cwiertny, D. M., Snyder, S. A., Schlenk, D. & Kolodziej, E. P. Environmental designer drugs: when transformation may not eliminate risk. *Environ. Sci. Technol.* 48, 11737–11745 (2014).
- Westerhoff, P., Yoon, Y., Snyder, S. & Wert, E. Fate of endocrine-disruptor, pharmaceutical, and personal care product chemicals during simulated drinking water treatment processes. *Environ. Sci. Technol.* 39, 6649–6663 (2005).
- Liu, Z. H., Kanjo, Y. & Mizutani, S. Removal mechanisms for endocrine disrupting compounds (EDCs) in wastewater treatment—physical means, biodegradation, and chemical advanced oxidation: a review. Sci. Total Environ. 407, 731–748 (2009).
- 28. Dijkstra, E. W. A note on two problems in connexion with graphs. *Numer. Math.* **1**, 269–271 (1959).
- Rice, J. & Westerhoff, P. Spatial and temporal variation in de facto wastewater reuse in drinking water systems across the USA. *Environ. Sci. Technol.* 49, 982–989 (2015)
- Rice, J. & Westerhoff, P. High levels of endocrine pollutants in US streams during low flow due to insufficient wastewater dilution. *Nat. Geosci.* 10, 587 (2017).
- Dickenson, E. R. V., Snyder, S. A., Sedlak, D. L. & Drewes, J. E. Indicator compounds for assessment of wastewater effluent contributions to flow and water quality. Water Res. 45, 1199–1212 (2011).
- Muñoz, I. et al. Ranking potential impacts of priority and emerging pollutants in urban wastewater through life cycle impact assessment. Chemosphere 74, 37–44 (2008).
- Morrall, D. et al. A field study of triclosan loss rates in river water (Cibolo Creek, TX). Chemosphere 54, 653–660 (2004).
- Glassmeyer, S. T. et al. Nationwide reconnaissance of contaminants of emerging concern in source and treated drinking waters of the United States. Sci. Total Environ. 581, 909–922 (2017).
- Gerrity, D. et al. Pilot-scale evaluation of ozone and biological activated carbon for trace organic contaminant mitigation and disinfection. Water Res. 45, 2155–2165 (2011).
- 36. Gerrity, D., Pecson, B., Trussell, R. S. & Trussell, R. R. Potable reuse treatment trains throughout the world. *J. Water Supply. Res. Technol. AQUA* **62**, 321 (2013).
- Hollender, J. et al. Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. *Environ. Sci. Technol.* 43, 7862–7869 (2009).
- Heberer, T. Tracking persistent pharmaceutical residues from municipal sewage to drinking water. J. Hydrol. 266, 175–189 (2002).
- 39. Kim, S. et al. Removal of contaminants of emerging concern by membranes in water and wastewater: a review. *Chem. Eng. J.* **335**, 896–914 (2017).
- Sophia, A. C. & Lima, E. C. Removal of emerging contaminants from the environment by adsorption. Ecotox. Environ. Safe. 150, 1–17 (2018).
- Scheurer, M., Storck, F. R., Brauch, H. J. & Lange, F. T. Performance of conventional multi-barrier drinking water treatment plants for the removal of four artificial sweeteners. Water Res. 44, 3573–3584 (2010).
- Wols, B. A. & Hofman-Caris, C. H. M. Review of photochemical reaction constants of organic micropollutants required for UV advanced oxidation processes in water. Water Res. 46, 2815–2827 (2012).
- Yu, Z. R., Peldszus, S. & Huck, P. M. Adsorption characteristics of selected pharmaceuticals and an endocrine disrupting compound—naproxen, carbamazepine and nonylphenol—on activated carbon. Water Res. 42, 2873–2882 (2008).
- Allaire, M., Wu, H. & Lall, U. National trends in drinking water quality violations. Proc. Natl Acad. Sci. 6, 2078–2083 (2018).
- DeFelice, N. B., Leker, H. G. & Gibson, J. M. Annual cancer risks from chemicals in North Carolina community water systems. *Hum. Ecol. Risk Assess.* 23, 974–991 (2017).
- Zhen, H. J. et al. Assessing the impact of wastewater treatment plant effluent on downstream drinking water-source quality using a zebrafish (Danio Rerio) liver cell-based metabolomics approach. Water Res. 145, 198–209 (2018).
- Karakurt, S., Schmid, L., Hubner, U. & Drewes, J. E. Dynamics of wastewater effluent contributions in streams and impacts on drinking water supply via riverbank filtration in Germany—a National Reconnaissance. *Environ. Sci. Technol.* 53, 6154–6161 (2019).
- Wang, Z. M., Shao, D. G. & Westerhoff, P. Wastewater discharge impact on drinking water sources along the Yangtze River (China). Sci. Total Environ. 599, 1399–1407 (2017).
- Hristovski, K. D., Pacemska-Atanasova, T., Olson, L. W., Markovski, J. & Mitev, T. Potential health implications of water resources depletion and sewage discharges in the Republic of Macedonia. J. Water Health 14, 682–691 (2016).
- Kapo, K. E. et al. iSTREEM (R): an approach for broad-scale in-stream exposure assessment of "down-the-drain" chemicals. *Integr. Environ. Assess. Manag.* 12, 782–792 (2016).
- 51. Loos, R. et al. EU-wide survey of polar organic persistent pollutants in European river waters. *Environ. Pollut.* **157**, 561–568 (2009).



- 52. Hass, U., Duennbier, U. & Massmann, G. Occurrence and distribution of psychoactive compounds and their metabolites in the urban water cycle of Berlin (Germany). *Water Res.* **46**, 6013–6022 (2012).
- Simazaki, D. et al. Occurrence of selected pharmaceuticals at drinking water purification plants in Japan and implications for human health. Water Res. 76, 187–200 (2015).
- 54. TWDB. State Water Plan: Water for Texas 150 (Texas Water Development Board, 2017).
- Stillwell, A. S., King, C. W., Webber, M. E., Duncan, I. J. & Hardberger, A. The energywater nexus in Texas. *Ecol. Soc.* 16, 2 (2011). http://www.ecologyandsociety.org/ vol16/iss1/art2/.
- Strahler, A. A quantitative analysis of watershed geomorphology. *Trans. Am. Geophys. Union* 38, 913–920 (1957).
- 57. Horton, R. E. Erosional development of streams and their drainage basins: hydrophysical approach to quantitative morphology. *Geol. Soc. Am. Bull.* **56**, 275–370 (1945).
- Pierson, S. M., Rosenbaum, B. J., McKay, L. D. & Dewald, T. G. Strahler Stream Order and Strahler Calculator Values in NHDPLUS. SOSC Technical Paper 11 (United States Geological Survey, 2008).

- Quality, T. C. o. E. in 2010 Texas Surface Water Quality Standards 1–217 (2010).
  Fono, L. J., Kolodziej, E. P. & Sedlak, D. L. Attenuation of wastewater-derived contaminants in an effluent-dominated river. Environ. Sci. Technol. 40, 7257–7262 (2006).
- Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2019