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Nationwide reconnaissance of contaminants of emerging concern in source and treated drinking waters of the United States

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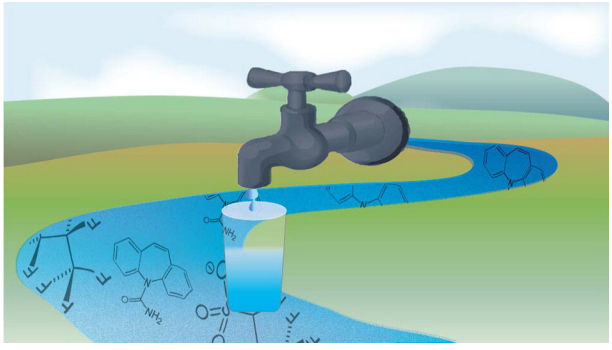
HIGHLIGHTS

- Nationwide study of 29 paired source water and treated drinking water samples
- Chemicals: pharmaceuticals, PFASs, anthropogenic waste indicators, and inorganics
- Microorganisms: bacteria, fungi, protozoa and viruses
- 148 contaminants detected in source water; 121 detected in treated drinking water.
- Provides a baseline for future drinking water monitoring for these constituents

GRAPHICAL ABSTRACT



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ABSTRACT

When chemical or microbial contaminants are assessed for potential effect or possible regulation in ambient and drinking waters, a critical first step is determining if the contaminants occur and if they are at concentrations that may cause human or ecological health concerns. To this end, source and treated drinking water samples from 29 drinking water treatment plants (DWTPs) were analyzed as part of a two-phase study to determine whether chemical and microbial constituents, many of which are considered contaminants of emerging concern, were

Abbreviations: AWI, anthropogenic waste indicator; CEC, contaminant of emerging concern; CCL, Contaminant Candidate List; DWTP, drinking water treatment plant; LCMRL, lowest concentration minimum reporting level; PFAS, per- and polyfluoroalkyl substances; PWS, public water system; QA/QC, quality assurance/quality control; RL, reporting level; SDWA, Safe Drinking Water Act; UCMR, Unregulated Contaminant Monitoring Rule; USEPA, United States Environmental Protection Agency; USGS, United States Geological Survey.

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detectable in the waters. Of the 84 chemicals monitored in the 9 Phase I DWTPs, 27 were detected at least once in the source water, and 21 were detected at least once in treated drinking water. In Phase II, which was a broader and more comprehensive assessment, 247 chemical and microbial analytes were measured in 25 DWTPs, with 148 detected at least once in the source water, and 121 detected at least once in the treated drinking water. The frequency of detection was often related to the analyte's contaminant class, as pharmaceuticals and anthropogenic waste indicators tended to be infrequently detected and more easily removed during treatment, while per and polyfluoroalkyl substances and inorganic constituents were both more frequently detected and, overall, more resistant to treatment. The data collected as part of this project will be used to help inform evaluation of unregulated contaminants in surface water, groundwater, and drinking water.

1. Introduction

There is increasing public concern over the detection of chemicals in water whose presence results from the diverse array of frequently used consumer, health-, and personal-care products. Chemicals contained in these products— including pharmaceuticals, fragrances, surfactants, and pesticides— may be present in wastewater influent through excretion, bathing, or direct disposal. Many of these chemicals have been documented to survive wastewater treatment and be discharged to surface and groundwaters. Previous reviews (Halling-Sorensen et al., 1998; Daughton and Ternes, 1999; Heberer, 2002; Diaz-Cruz and Barcelo, 2004; Glassmeyer et al., 2008; Kostich et al., 2010; Delgado et al., 2012; Pal et al., 2014; Li et al., 2015; Petrie et al., 2015) have summarized the peer-reviewed literature reporting the occurrences of these chemicals in water resources. Initially termed "emerging contaminants", there is some misperception that the term suggests that these chemicals have only recently been released into the environment. In fact, these chemicals have been released as long as they have been in use, and some compounds (such as caffeine) have been detected in wastewater (Shuval and Gruener, 1973; Shackelford and Cline, 1986), surface water (Donaldson, 1977; Sheldon and Hites, 1978; Eganhouse et al., 1983; Richardson and Bowron, 1985), and drinking water (Coleman et al., 1980) for several decades. What is emerging is greater awareness by the general public of the presence of these contaminants in the environment and the direct link of environmental presence to household use. The ability of environmental scientists to detect extremely low ambient concentrations of these contaminants, aided by improvements to the analytical instrumentation, further fosters this awareness. Thus, the term "contaminants of emerging concern" (CECs) is a more appropriate choice when describing these contaminants in aggregate.

In the United States, the Safe Drinking Water Act (SDWA), as amended in 1996 (USEPA, 1996) gives the US Environmental Protection Agency (USEPA) the authority to regulate contaminants in finished drinking water, as well as to protect drinking water sources. To regulate a contaminant in drinking water, the SDWA requires that three criteria must be met: 1) the contaminant may have an adverse effect on the health of persons, 2) the contaminant is known to occur or there is a substantial likelihood the contaminant will occur in drinking water with a frequency and at levels of public health concern, and 3) in the sole judgment of the USEPA Administrator, regulation of the contaminant presents a meaningful opportunity for reducing health risks for persons served by public water systems. The SDWA requires the USEPA to evaluate unregulated chemical and microbial contaminants which may necessitate future regulation through the Contaminant Candidate List (CCL) process; the draft fourth CCL (CCL 4) was proposed in 2015 (USEPA, 2015). Whether a contaminant is known or anticipated to occur in public water systems is considered as part of the CCL process, along with potential health effects.

Compared to other environmental matrices, there are a paucity of studies that have assessed occurrence of CECs in finished drinking water (Benotti et al., 2009; Stackelberg et al., 2004; Stackelberg et al., 2007; Snyder, 2008; Garcia-Ac et al., 2009; Loos et al., 2007; Togola and Budzinski, 2007), and these studies typically do not examine analytes from multiple contaminant classes. One mechanism to obtain nationally representative drinking water occurrence data is through the Unregulated Contaminant Monitoring Regulation (UCMR), an authority that allows the USEPA to gather occurrence data from all public water systems (PWS) serving >10,000 people, and a representative sample of PWSs serving 10,000 or fewer people, for no more than 30 contaminants in five-year cycles (USEPA, 2012a). Occurrence data of CECs in drinking water in published studies helps determine which analytes would be most appropriate for the UCMR. However, focused, national-scale studies of CEC presence and concentration in source- and treated drinking water samples that use consistent, state-of-the-art sample collection and analysis approaches and assessing the widest array of CECs offer the greatest benefit for identifying the most appropriate contaminants for any detailed UCMR assessments.

This paper is one of a series of papers describing a comprehensive study on the presence, concentrations, and persistence of chemical and microbial CECs in source and treated drinking waters of the United States (Batt et al., 2016; Benson et al., 2016; Conley et al., 2016; Furlong et al., 2016; King et al., 2016; Kostich et al., 2016; Boone et al., unpublished results; Varughese et al., unpublished results). This was a joint effort of the USEPA and the U.S. Geological Survey (USGS), as part of a long-term interagency agreement. A primary goal of the overall study was to provide accurate, objective information for assessing the potential for human exposure to a comprehensive set of CECs via drinking water. A secondary goal was to evaluate removal, if any, of CECs from source waters by currently used drinking water treatment processes under typical plant operating conditions. The interdisciplinary approach of this nationwide study is unique in that it combined both the measurement of CECs along with the evaluation of the potential effects of the contaminants, through both an in vitro estrogenic activity bioassay and screening level human and ecological health impact assessments.

2. Experimental design

This study was conducted in two phases. In Phase I (2007), source and treated drinking water from nine drinking water treatment plants (DWTPs) from eight states across the United States were sampled and analyzed for 84 chemicals using three different analytical methods. The Phase I effort provided an opportunity to test the experimental design, field sampling protocols, and analytical methods as applied to operator-collected samples from DWTPs. In Phase II (2010—2012), the quality assurance/quality control design was refined, the analyte list expanded (247 chemical and microbiological contaminants using 16 different methods, as well as an in vitro estrogenicity bioassay), and the number of DWTPs sampled increased to 25 DWTPs located in 24 states, including five that were also sampled in Phase I. Between the two phases, 29 DWTPs were investigated (five in both Phase I and II, four in Phase I only and 20 in Phase II only). A total of 77 common analytes were measured in both Phase I and II.

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