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1,4-Dioxane drinking water occurrence data from the third unregulated contaminant monitoring rule

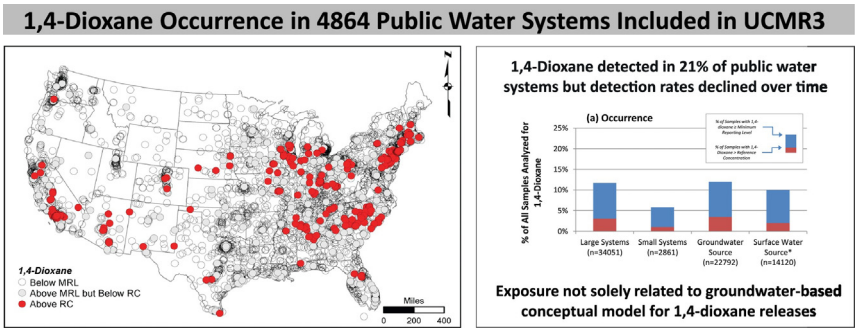
David T. Adamson [a,*], Elizabeth A. Piña [a], Abigail E. Cartwright [b], Sharon R. Rauch [a], R. Hunter Anderson [c], Thomas Mohr [d], John A. Connor [a]

[a] GSI Environmental Inc., Houston, TX 77098, USA
[b] Rice University, 6100 Main Street, Houston, TX 77005, USA
[c] Air Force Civil Engineer Center, San Antonio, TX 78226, USA
[d] Santa Clara Valley Water District, 5750 Almaden Expressway, San Jose, CA 95118, USA

HIGHLIGHTS

- 1,4-Dioxane and other unregulated contaminants in drinking water were evaluated.
- 1,4-Dioxane exhibited relatively high rates of detection in public water systems.
- 1,4-Dioxane did not follow common assumptions about release and exposure routes.
- Regulatory determinations on 1,4-dioxane will have significant implications.

GRAPHICAL ABSTRACT



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ABSTRACT

This study examined data collected from U.S. public drinking water supplies in support of the recently-completed third round of the Unregulated Contaminant Monitoring Rule (UCMR3) to better understand the nature and occurrence of 1,4-dioxane and the basis for establishing drinking water standards. The purpose was to evaluate whether the occurrence data for this emerging but federally-unregulated contaminant fit with common conceptual models, including its persistence and the importance of groundwater contamination for potential exposure. 1,4-Dioxane was detected in samples from 21% of 4864 PWSs, and was in exceedance of the health-based reference concentration (0.35 μg/L) at 6.9% of these systems. In both measures, it ranked second among the 28 UCMR3 contaminants. Although much of the focus on 1,4-dioxane has been its role as a groundwater contaminant, the detection frequency for 1,4-dioxane in surface water was only marginally lower than in groundwater (by a factor of 1.25; $p < 0.0001$). However, groundwater concentrations were higher than those in surface water ($p < 0.0001$) and contributed to a higher frequency of exceeding the reference concentration (by a factor of 1.8, $p < 0.0001$), indicating that surface water sources tend to be more dilute. Sampling from large systems increased the likelihood that 1,4-dioxane was detected by a factor of 2.18 times relative to small systems ($p < 0.0001$). 1,4-Dioxane detections in drinking water were highly associated with detections of other chlorinated compounds particularly 1,1-dichlorethane (odds ratio = 47; $p < 0.0001$), which is associated with the release of 1,4-dioxane as a chlorinated solvent stabilizer. Based on aggregated nationwide data, 1,4-dioxane showed evidence of a decreasing trend in concentration and detection frequency over time. These data suggest that the loading to drinking

* Corresponding author at: GSI Environmental Inc., 2211 Norfolk, Suite 1000, Houston, TX 77098, USA.
E-mail address: dtadamson@gsi-net.com (D.T. Adamson).

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water supplies may be decreasing. However, in the interim, some water supply systems may need to consider improving their treatment capabilities in response to further regulatory review of this compound.

1. Introduction

The fate and transport of 1,4-dioxane as an emerging environmental contaminant is an area of considerable interest (Mohr et al., 2010). USEPA recently included 1,4-dioxane on its initial list of 10 "high priority" chemical substances as part of the 2016 amendments to the Toxic Substances Control Act. Based on toxicological data, it is classified as "likely to be carcinogenic to humans" by USEPA, with ingestion of drinking water identified as a potential exposure pathway, Many states have adopted screening levels or cleanup standards for 1,4-dioxane in water, but these levels vary over an order of magnitude (Suthersan et al., 2016). Currently, there is no federally-enforceable drinking water standard for this compound. In part, this is due to a lack of understanding of its prevalence and exposure potential (Richardson and Kimura, 2016; Suthersan et al., 2016). For contaminants like 1,4-dioxane that are not yet regulated in drinking water, USEPA has developed a systematic process for evaluating individual chemicals to support a definitive regulatory determination that ensures that drinking water ingestion poses no significant risk to the general public. This process relies on integrating information on the toxicological characteristics of individual compounds, the potential for exposure (i.e., occurrence), and the extent of risk reduction that would occur if the compound was regulated. To a lesser extent, the technical challenges associated with preventing exposure are also considered when determining the appropriate concentration limit. To achieve this mandate with the necessary level of confidence, there must a significant amount of data to evaluate. A key step in this process is the development of a Candidate Contaminant List (CCL) that is periodically reviewed and updated. The CCL is determined based on the results of a series of formal monitoring programs that are mandated through the Unregulated Contaminant Monitoring Rule (UCMR). The UCMR is designed to collect occurrence data from a pre-determined number of public water systems (PWSs) as part of a 5-year review cycle, and USEPA has completed two rounds of UCMR (UCMR1, with monitoring between 2001 and 2003; UCMR2, with monitoring between 2008 and 2010).

UCMR3 monitoring was completed in December 2015 and represented the latest effort at supporting this regulatory process. 1,4-Dioxane was one of 28 unregulated contaminants that were monitored during the three-year reporting period for UCMR3 (note that 1,4-dioxane was not included in UCMR1 or UCMR2). Based on pre-existing data, many of the UCMR3 contaminants were expected to have a higher frequency of occurrence than seen in earlier rounds of UCMR monitoring. This expectation was partly based on the low minimum reporting levels (MRLs) that were required as part of UCMR3; for most contaminants, these MRLs are <1 μg/L. The inclusion of mandated MRLs that were established by screening lab and analytical capabilities was a key difference for UCMR3 relative to UCMR1 and UCMR2. Another critical component for UCMR3 (similar to UCMR1 and UCMR2) was the use of a "reference concentration" (RC) for the majority of contaminants on the monitoring list. These serve as health-based reference levels and are based on the best-available toxicological information at the time the rule was developed (2012).

The complete UCMR3 dataset is now available for review, but a full regulatory evaluation of the results will take several years due to the size of the dataset as well as the general prudence associated with the CCL review process (e.g., USEPA published reviews of UCMR1 and UCMR2 data four to five years after the end of the respective monitoring periods (USEPA, 2008, 2015). Consequently, there is an opportunity to present a timelier and thorough review of this newly-completed UCMR3 dataset to better serve environmental professionals, the regulatory community, and the public at large. Several recent studies highlight the utility of the UCMR3 data for analyzing contaminant trends. For example, Hu et al. (2016) reported occurrence rates for poly- and perfluoroalkyl substances (PFASs) and attempted to quantify the relative contribution of potential sources to the PFAS detections in public water supplies. Chebeir et al. (2016) summarized chromium distribution UCMR3 data and showed that significant oxidation of trivalent chromium to hexavalent chromium was occurring within the drinking water distribution systems.

The study described herein focused on a detailed evaluation of 1,4-dioxane as it is among the most frequently detected contaminants in UCMR3. 1,4-Dioxane is an anthropogenic compound highly prevalent in groundwater at industrial sites contaminated with chlorinated solvents, in particular 1,1,1-trichloroethane, due to its historic use as a stabilizer (Mohr et al., 2010; Anderson et al., 2012; Adamson et al., 2014). As a result, these subsurface releases of 1,4-dioxane to groundwater represent a critical concern for the Department of Defense and other stakeholders (Anderson et al., 2012). These concerns are exacerbated by the technical challenges associated with removing 1,4-dioxane from water using either in situ (e.g., injection of remedial amendments) or ex situ (e.g., conventional or advanced drinking water treatment) technologies (Zenker et al., 2003; USEPA, 2006, 2014; Stepien et al., 2014). 1,4-Dioxane has also shown widespread occurrence in wastewater treatment plant effluents (Simonich et al., 2013), meaning that discharges to surface water represent a potential risk to downstream drinking water treatment plants. However, these discharges to surface water are diluted in the receiving stream, and there is empirical evidence suggesting that dilution of 1,4-dioxane should largely mitigate any impacts to drinking water supplied from surface water (Simonich et al., 2013).

These issues have contributed to a significant debate about appropriate health-based drinking water standards for 1,4-dioxane, and several states have set standards below those proposed by USEPA (USEPA, 2014; MADEP, 2015; Suthersan et al., 2016). Since UCMR3 required that PWSs report information on individual sample concentrations, system size, and source water types, the database also provides a means to better understand the nature of 1,4-dioxane occurrence and whether it fits our common conceptual model for 1,4-dioxane sources and behavior in the environment. This includes the assumptions that potential 1,4-dioxane exposures are largely associated with groundwater, where the compound co-occurs with chlorinated solvents and is persistent. The objectives of this study were to (1) quantify the relative occurrence of 1,4-dioxane in drinking water supplies to better understand the impact of future regulatory determinations; (2) establish the relative importance of certain system characteristics and co-contaminants on the extent and magnitude of 1,4-dioxane detections; and (3) evaluate if there is evidence for 1,4-dioxane attenuation over time. These data have tremendous technical and cost implications for stakeholders because they highlight the potential effect of increased regulations and identify where resources may have to be allocated to address these effects.

2. Methods

2.1. Data sources

Data were obtained from the National Contaminant Occurrence Database (NCOD) maintained by the USEPA for the purposes of tracking UCMR3 and similar occurrence data (USEPA, 2016b). UCMR3 monitoring began in January 2013 and included 28 contaminants (see Fig. 1).