Review of perchlorate occurrence in large public drinking water systems in the United States of America

Steven J. Luis, Elizabeth A. Miesner, Clarissa L. Enslin and Keith Heidecorn

ABSTRACT

When deciding whether or not to regulate a chemical, regulatory bodies often evaluate the degree to which the public may be exposed by evaluating the chemical's occurrence in food and drinking water. As part of its decision-making process, the United States Environmental Protection Agency (USEPA) evaluated the occurrence of perchlorate in public drinking water by sampling public water systems (PWSs) as part of the first implementation of the Unregulated Contaminant Monitoring Rule (UCMR 1) between 2001 and 2005. The objective of this paper is to evaluate the current representativeness of the UCMR 1 dataset. To achieve this objective, publicly available sources were searched to obtain updated perchlorate data for the majority of large PWSs with perchlorate detections under UCMR 1. Comparison of the updated and UCMR 1 perchlorate datasets shows that the UCMR 1 dataset is no longer representative because the extent and degree of occurrence has decreased since implementation of UCMR 1. Given this finding, it seems appropriate for regulatory bodies engaged in decision-making processes over several years to periodically re-evaluate the conditions that prompted the regulatory effort, thereby ensuring that rules and regulations address actual conditions of concern.

Key words | drinking water, groundwater, perchlorate occurrence, public water supply, Unregulated Contaminant Monitoring Rule

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INTRODUCTION

Perchlorate (ClO_4^-) has been detected in the environment as a result of both anthropogenic and natural sources. Perchlorate commonly occurs as one of the following salts: ammonium perchlorate (NH_3ClO_4), potassium perchlorate ($KClO_4$), and sodium perchlorate ($NaClO_4$). Ammonium perchlorate has been used as the primary oxidant in solid rocket propellants and explosives for several decades. Potassium and sodium perchlorate have also been used in a variety of explosives and other military applications. In addition to industrial sources, some consumer products contain perchlorate, including fireworks, flares, matches, and chlorine bleach (ITRC 2005).

Aside from perchlorate from anthropogenic sources, naturally-occurring perchlorate has also been detected in the environment. The presence of naturally-occurring perchlorate was first documented in the sodium nitrate deposits of Chile's Atacama Desert. Chilean nitrate deposits, sometimes referred to as 'nitrate caliche,' were widely used as fertilizer in the United States during the first half of the 20th century and thus constitute a significant non-anthropogenic source of perchlorate in the environment. Naturally occurring perchlorate has also been detected in groundwater in arid regions of the southwestern United States (Bao & Gu 2004; Jackson *et al.* 2006).

In addition to the United States and Chile, perchlorate has also been reported in the environment and/or food and water supplies in member states of the European Union, including France, Germany, and the United

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Kingdom, as well as in Japan, Korea, Canada, Bolivia, Israel, and Antarctica. (ITRC 2005; Kosaka *et al.* 2007; Quiñones *et al.* 2007; Gal *et al.* 2008; Kounaves *et al.* 2010; Asami *et al.* 2013; Bundesinstitut für Risikobewertung 2013; Agence Nationale de Sécurité Sanitaire Alimentation Environnement Travail 2014; Nawaz 2016). The presence of perchlorate in the environment is of concern because perchlorate can inhibit iodine uptake by the thyroid, thereby affecting thyroid function (NRC 2005).

As a consequence of perchlorate detections and concerns about potential human health effects, various government agencies have considered regulating perchlorate. As part of these regulatory decision-making processes, studies have been conducted to assess the degree to which the public may be exposed to perchlorate by evaluating the occurrence of perchlorate in food and/or drinking water. For example, occurrence studies have been conducted in Europe (Bundesinstitut für Risikobewertung 2013; Nawaz 2016; Arcella *et al.* 2017). This paper focuses on the dataset from one such perchlorate occurrence study conducted in the United States.

PERCHLORATE OCCURRENCE IN THE UNITED STATES AND THE SAFE DRINKING WATER ACT

To regulate a contaminant that may be present in drinking water under the Safe Drinking Water Act (SDWA), one of the considerations is the degree to which the contaminant occurs in drinking water. To assess the degree to which a contaminant is present in drinking water in the United States, on September 17, 1999, the United States Environmental Protection Agency (USEPA) promulgated the Unregulated Contaminant Monitoring Rule (UCMR) for Public Water Systems (PWSs) pursuant to the SDWA, as amended in 1996 (64 Fed Reg. 50556, September 17, 1999). The UCMR required all large community and non-transient non-community water systems serving more than 10,000 persons and 800 representative small systems serving 10,000 or fewer persons to monitor for contaminants selected from the USEPA's Contaminant Candidate List (CCL) developed in 1998. Generated data would be used to evaluate and prioritize contaminants on the CCL, support determinations to regulate contaminants, and support the development of drinking water regulations. The first implementation of the UCMR became known as UCMR 1. Perchlorate was included on the CCL and, thus, was included for monitoring in the UCMR 1.

UCMR 1 sampling was conducted between 2001 and 2005 and included all large (serving more than 10,000 persons) PWSs in the United States and a statistically representative sampling of small PWSs (i.e., those PWSs serving 10,000 or fewer persons) which were sampled under the USEPA's UCMR. A table summarizing the sampling results is available on USEPA's website (https://www.epa.gov/dwucmr/occurrence-data-unregulated-contaminant-monitoring-rule), but underlying laboratory reports and other supporting information are not provided.

Altogether, 3,068 large and 797 small PWSs were sampled under UCMR 1. Of the 3,068 large PWSs sampled, perchlorate was detected in 152 PWSs. Of the 797 small PWSs sampled, perchlorate was detected in 8 PWSs. Subsequent UCMR monitoring programs UCMR 2 and UCMR 3 were implemented in 2007–2011 and 2012–2016, respectively, but perchlorate was not monitored. As a result, the 152 large PWSs and 8 small PWSs in which perchlorate was detected under UCMR 1 were not subsequently re-sampled under the UCMR program.

In 2005, an analysis by the National Research Council (NRC) derived a perchlorate reference dose (RfD) of 0.0007 milligrams per kilogram-day (mg/kg-day). Using default exposure assumptions (e.g., drinking water ingestion rate of 2 liters per day), USEPA calculated a Drinking Water Equivalent Level (DWEL) of 24.5 μg/L (USEPA 2009).

In 2008, USEPA summarized its evaluation of UCMR 1 results in the Federal Register by comparing the sample results with the following threshold concentrations: 4, 5, 7, 10, 12, 15, 17, 20, and 25 μg/L. USEPA also summarized the US population associated with small and large PWSs within each threshold concentration range. In 2008, USEPA also arrived at the preliminary determination that perchlorate did not occur with a frequency and at levels of public health concern and that regulation was not warranted (73 Fed Reg. 60262, October 10, 2008). USEPA's summary is provided in Table 1.

In 2011, USEPA reversed its 2008 preliminary determination and decided to regulate perchlorate (76 Fed Reg. 7762, February 11, 2011). At the time of writing, USEPA had not finalized a perchlorate regulation.

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Table 1 | UCMR 1 occurrence and population estimates for perchlorate and comparison to various thresholds (73 Fed Reg. 198)

Threshold (μg/L)	PWSs with at least 1 detection > threshold	Population served by PWSs with at least 1 detection > threshold (Millions)
4	4.01% (155 out of 3,865)	16.6
5	3.16% (122 out of 3,865)	14.6
7	2.12% (82 out of 3,865)	7.2
10	1.35% (52 out of 3,865)	5
12	1.09% (42 out of 3,865)	3.6
15	0.80% (31 out of 3,865)	2
17	0.70% (27 out of 3,865)	1.9
20	0.49% (19 out of 3,865)	1.5
25	0.36% (14 out of 3,865)	1

PWSs – Public Water Systems.
UCMR 1 – Unregulated Contaminant Monitoring Rule 1.

USEPA's process for developing new regulations is appropriately thorough. However, as the case of perchlorate shows, the process has extended over several years. The case of perchlorate in the United States is consistent with the observation of Kucharzyk *et al.* (2009), who noted that USEPA's process of developing regulations can take years to complete.

CHANGES IN CONDITIONS SINCE EVALUATING PERCHLORATE DRINKING WATER OCCURRENCE UNDER UCMR 1

In the more than 10 years since completion of UCMR 1 in 2005, a number of changes in conditions have taken place that likely affect current perchlorate occurrence in drinking water (ITRC 2008; Russell & Morley 2017). Changes include the following:

- Promulgation of water quality guidelines and regulations, most notably California's Maximum Contaminant Level (MCL) of 6 micrograms per litre (μg/L) and Massachusetts' MCL of 2 μg/L.
- At many locations with groundwater and/or surface water perchlorate impacts, remedial and source control efforts have reduced the magnitude and spatial extent of those impacts. For example, USEPA reports that the rate of perchlorate entering Lake Mead decreased 90% between 1997 and 2016 due to ongoing cleanup activities (USEPA 2016a). Substantial progress has been reported for other perchlorate-impacted groundwater sites (see, for example, USEPA (2016b) and CalEPA DTSC (2017)).
- At some perchlorate-impacted groundwater sites, treated or alternative water supplies have been provided to local communities. These efforts have been underway for several years and have generally been successful in reducing the potential for perchlorate exposure (Russell & Morley 2017). For example, to ensure that perchlorate concentrations do not exceed 1 μg/L, the City of Aberdeen, Maryland has been managing operation of its drinking water wells, including the installation of perchlorate filtration units in 2005 (Aberdeen 2017).
- Analytical methods for perchlorate have improved since implementation of UCMR 1, reducing the likelihood of false positive detections of perchlorate and improving sensitivity (i.e., lowering detection limits), thereby improving the reliability of analytical results (Christen 2003; Department of Defense Environmental Data Quality Workgroup 2007; ITRC 2008).

Most of the changes listed above are self-explanatory. However, changes in analytical methods may require additional explanation. On March 2, 2000, USEPA Method 314.0 was specified for use in analyzing for perchlorate in UCMR 1 (65 Fed Reg. 11371, March 2, 2000). USEPA Method 314.0 was specified following promulgation of UCMR 1 on September 17, 1999 because USEPA had not validated the method at the time of promulgating UCMR 1. At the time of specifying USEPA Method 314.0 for use in UCMR 1 perchlorate sampling, USEPA also specified procedures for laboratories to receive approval for use of the new method. Under UCMR 1, samples were analyzed using USEPA Method 314.0 with a Method Reporting Limit (MRL) of 4.0 μg/L.

Subsequent to publication, USEPA Method 314.0 has come to be recognized as resulting in false detections and elevated concentrations under some circumstances due to the method's reliance on ion chromatography. Such circumstances include interferences and loss of sensitivity in

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sample matrices with relatively high total dissolved solids (TDS) and high concentrations of common anions such as chloride, sulfate, and carbonate. (CalEPA DTSC 2004; Quiñones *et al.* 2007; ITRC 2008; Kucharzyk *et al.* 2009).

Analytical issues associated with USEPA Method 314.0 have been noted by workers at a number of perchlorate-impacted hazardous waste sites. Although seemingly minor, concerns about the reliability of analytical methods are understandable in light of the relatively low concentrations at which USEPA has considered regulating perchlorate, as reflected in Table 1, and California and Massachusetts MCLs of 6 μg/L and 2 μg/L, respectively. DTSC and the Department of Defense have issued guidance advising of these issues and recommending steps to increase the reliability of perchlorate detections (CalEPA DTSC 2004; Department of Defense Environmental Data Quality Workgroup 2007).

As indicated by one of the developers of USEPA Method 314.0, limitations of the method have led to a search for improved methods for perchlorate analysis (Christen 2003). Since publication of USEPA Method 314.0, other analytical methods for perchlorate have been developed, including USEPA Methods 314.1 and 331.0. The newer analytical methods improve upon and address shortcomings of USEPA Method 314.0, including those related to interference and loss of sensitivity (ITRC 2008). As noted above, the improved methods were not available at the time UCMR 1 sampling was conducted.

UCMR 1 DRINKING WATER SAMPLING METHODS, RESULTS, AND PREVIOUS REVIEWS

Under UCMR 1, PWSs with surface water sources were to be sampled quarterly over a one-year period and PWSs with groundwater sources were to be sampled semi-annually over a one-year period. Actual sampling practices varied considerably, with significantly larger numbers of samples collected for some PWSs than for other PWSs (Brandhuber *et al.* 2009).

UCMR 1 sampling results for the 152 large PWSs using median, 90th percentile, and maximum perchlorate concentrations, the same summary statistics used by previous researchers such as AWWA (2008), are listed in Table 3. (Consistent with previous researchers evaluating the UCMR 1 dataset (e.g., Brandhuber *et al.* 2009), non-detects were set to half the detection limit, 2 μg/L). Table 3 also lists the 2005 US population associated with the respective PWSs in each concentration category. Figure 1 shows the distribution of UCMR 1 perchlorate concentrations and the 2005 US population potentially exposed to perchlorate, respectively, using the same percentiles as Table 3.

Additional UCMR 1 summary statistics are as follows:

- Perchlorate was detected in 647 out of 34,728 samples collected from small and large PWSs under UCMR 1, for a total of 160 PWSs with perchlorate detections. Out of 5,687 samples collected from the 152 large PWSs with perchlorate detections, perchlorate was detected in 632 samples, an overall detection rate of approximately 11%.
- Perchlorate concentrations ranged from a minimum of 4 μg/L (the MRL at the time of UCMR 1 implementation) to a maximum of 420 μg/L.
- The frequency of perchlorate detection for a given PWS ranged from 0.61% (1 detection out of 162 samples, e.g., Public Water System Identification Code (PWSID) CA3310001– Coachella Valley Water District (VWD): Cove Community) to 100% (8 detections out of 8 samples, e.g., PWSID NV0000289 – Southern Nevada Water System).

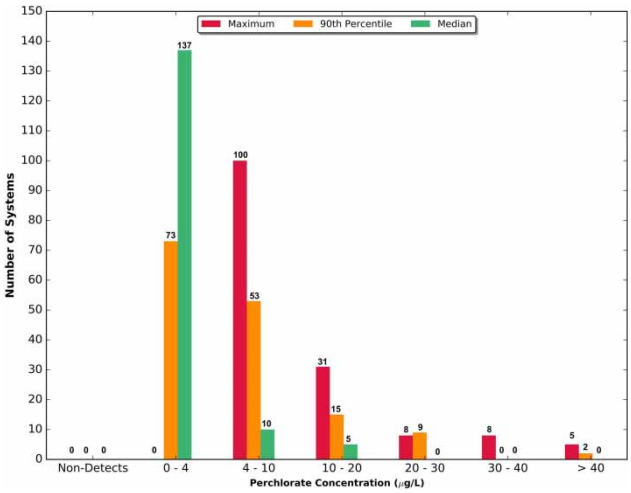

Figure 1 | Distribution of perchlorate sample results for 152 large PWSs with perchlorate detections under UCMR 1.

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- 85% of the 152 large PWSs exhibiting a frequency of detection of less than 35%.
- Of the 152 large PWSs with perchlorate detections, almost 50% (73 PWSs) had only one perchlorate detection (i.e., the remaining samples for the PWS were non-detect).

PWSs in all 50 states and U.S. Territories were sampled under UCMR 1. Figures 2–4 show the locations of the 152 large PWSs in which perchlorate was detected as categorized by maximum, 90th percentile and median detections. Figures 2–4 show that perchlorate was detected at least once in 26 states and U.S. Territories.

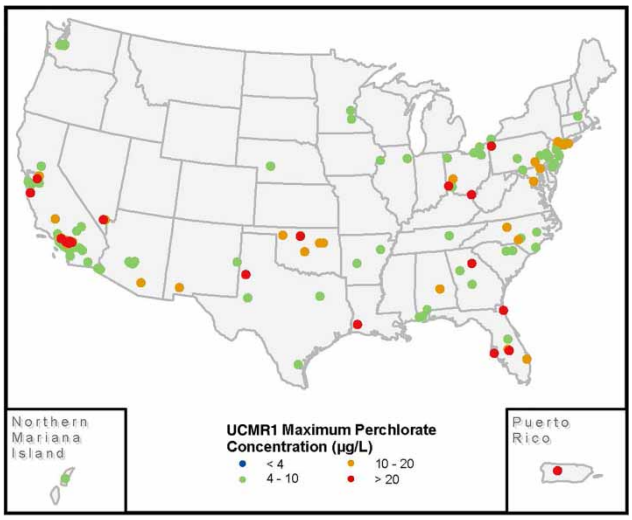

Figure 2 | UCMR 1 maximum perchlorate detections for large PWSs.

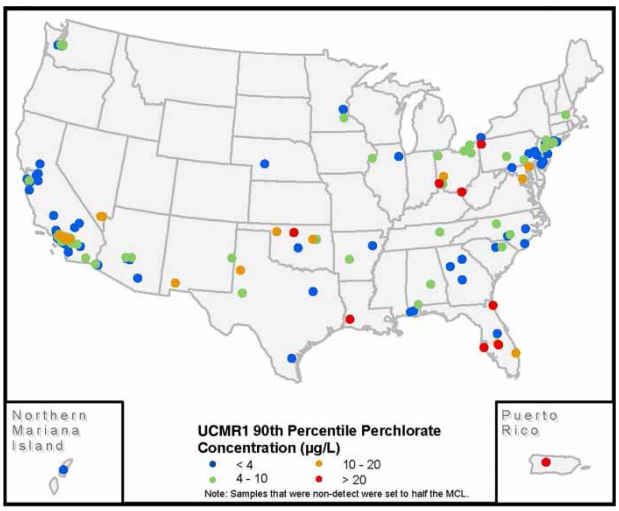

Figure 3 | UCMR 1 90th percentile perchlorate detections for large PWSs.

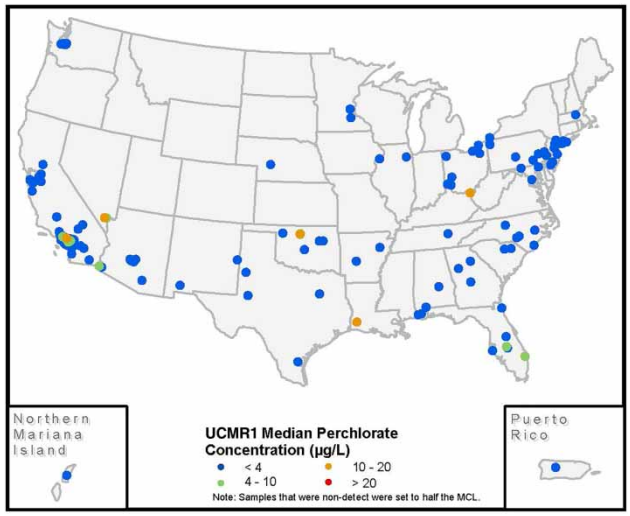

Figure 4 | UCMR 1 median perchlorate detections for large PWSs.

Previous reviewers have reported on the occurrence of perchlorate in drinking water in the United States. Previous publications are summarized in Table 2. As Table 2 shows, previous researchers focused on two issues: (1) interpretation of UCMR 1 data as well as data from more limited contemporaneous studies; and (2) estimation of costs associated with drinking water treatment to meet potential regulatory thresholds. The previous reviews listed in Table 2 differ from the present review in that they do not focus on the representativeness of the UCMR 1 perchlorate occurrence dataset and do not present an updated perchlorate occurrence dataset.

DEVELOPMENT OF AN UPDATED PERCHLORATE DRINKING WATER OCCURRENCE DATASET

Consideration of the changes in conditions listed above suggests that the UCMR 1 perchlorate dataset may no longer be representative of perchlorate occurrence in the United States. Given the potential for changes in perchlorate concentrations since UCMR 1, the objective of this review is to assess the current representativeness of the

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Table 2 | Summary of previous research

Year	Author	Article title	Summary
2004	Arizona Department of Environmental Quality	Perchlorate in Arizona Occurrence Study of 2004	Arizona Department of Environmental Quality performed state-wide sampling of surface and groundwater, and source water to evaluate the extent of perchlorate occurrence in Arizona. Summary maps of sampling results were developed showing perchlorate occurrence in PWSs in Arizona.
2004	Jackson *et al.*	Distribution and Potential Sources of Perchlorate in the High Plains Region of Texas	Jackson *et al.* performed sampling of 560 PWSs and 186 wells across 54 counties in northwest Texas to determine the source and distribution of perchlorate in groundwater. Results showed that perchlorate was detected at relatively low concentrations in the study area. The authors suggested that the presence of perchlorate is most likely due to atmospheric production and/or surface oxidative weathering in the region.
2004	Kennedy/Jenks	Cost of Compliance for Three Potential Perchlorate MCLs	Kennedy/Jenks developed cost estimates for PWSs to comply with potential MCLs. Kennedy/Jenk's analysis relied upon a water quality database obtained from the California Department of Health Services in July 2003.
2005	Brandhuber & Clark	Perchlorate Occurrence Mapping	Brandhuber & Clark analyzed preliminary UCMR 1 data available in late 2004 as well as data from studies conducted by the states of Arizona, California, Massachusetts, and Texas. Summary statistics and maps were developed showing the occurrence of perchlorate in PWSs across the United States.
2007	MassDEP	Evaluation of Perchlorate Contamination at a Fireworks Display	MassDEP performed several studies to determine the extent of perchlorate in surface water and groundwater in Massachusetts. Identified sources of perchlorate included blasting agents, military munitions, fireworks, and industrial perchloric acid use.
2008	AWWA	National Cost Implications of a Potential Perchlorate Regulation	AWWA performed a screening-level cost assessment to evaluate the cost implications of potential national regulatory levels for perchlorate in drinking water. AWWA's analysis was based on the UCMR 1 dataset. AWWA also took steps to verify the representativeness of the UCMR 1 dataset, including contacting some PWSs to verify estimated values.
2009	Brandhuber *et al.*	A Review of Perchlorate Occurrence in Public Drinking Water Systems	Brandhuber *et al.* analyzed PWS perchlorate occurrence data from UCMR 1 as well as data from studies conducted by the states of Arizona, California, Massachusetts, and Texas. Analyses included summary statistics and mapping of perchlorate occurrence in PWSs across the United States.
2009	Russell *et al.*	National cost implications of a perchlorate regulation	Russell *et al.* developed cost estimates for PWSs to comply with potential MCLs. The analysis relied upon the UCMR 1 dataset and focused on costs associated with source water treatment.
2017	California Division of Drinking Water (CDDW)	History of Perchlorate in California Drinking Water	The CDDW monitors perchlorate concentrations in hundreds of wells throughout the state of California. Monitoring commenced in 1997. Results are available for more than 7,000 drinking water sources.
2017	Russell & Morley	Estimating the National Costs of Regulating Perchlorate in Drinking Water	Russell & Morley developed cost estimates for PWSs to comply with potential MCLs. Compliance strategies to address perchlorate included source water treatment, source abandonment, and blending. Like Russell *et al.* (2009), the analysis relied up on the UCMR 1 perchlorate dataset. Perchlorate data from California and Massachusetts were also used.

PWSs – Public Water Systems.
UCMR 1 – Unregulated Contaminant Monitoring Rule 1.

UCMR 1 perchlorate dataset for those large PWSs with perchlorate detections under UCMR 1 and to update the dataset, as appropriate. It was not possible to sample the approximately 3,000 large PWSs in the United States, so it was necessary to identify an appropriate subset of PWSs for the purpose of this review. The focus of this review was on the 152 large PWSs that had perchlorate detections under UCMR 1 for the following reasons:

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Table 3 | United States population served by large PWSs with at least one perchlorate detection under UCMR 1

	Population served by PWS (Total population = 13,171,000)					
	Maximum		90th Percentile		Median	
Perchlorate threshold (μg/L)	Population	Percentage PWSs with at least 1 detection greater than threshold	Population	Percentage PWSs with at least 1 detection greater than threshold	Population	Percentage PWSs with at least 1 detection greater than threshold
0–4	0	0	8,523,000	65	12,167,000	92
4–10	8,386,000	64	3,248,000	25	665,000	5.0
10–20	3,264,000	25	486,000	3.7	339,000	3
20–30	584,000	4.4	850,000	6.5	0	0.0
30–40	502,000	3.8	0	0.0	0	0.0
>40	434,000	3.3	65,000	0.5	0	0.0

Notes:
1. Non-detects were set to half the detection limit, 2 μg/L.
2. Population numbers have been rounded to the nearest thousand.
3. Source of population data: EPA SDWIS Federal Reports. 2005 population data were not available for the following two PWSs: Southern Nevada Water System (PWSID NV 0000289) and Creek County Rural Water District, No. 1 (OK 1020419). 2017 population data were used for these PWSs.
4. Population data from the Metropolitan Water District of Southern California (MWD) were excluded from the above table to avoid double counting. MWD supplies water to many PWSs Southern California, including some PWSs under consideration here.
PWSs – Public Water Systems.
UCMR 1 – Unregulated Contaminant Monitoring Rule 1.

- UCMR 1 sampling of large PWSs was comprehensive. Unlike small PWSs, all large PWSs were sampled under UCMR 1. Therefore, the 152 large PWSs with perchlorate detections, approximately 5% of all large PWSs, constitute the entire population of large PWSs with perchlorate detections under UCMR 1.
- As noted by Brandhuber *et al.* (2009), a stratified sampling strategy was employed by USEPA for small PWSs. Without the weighting factors used for each stratum, it is not possible to extend UCMR 1 sample results to all small PWSs in a statistically valid manner.
- Altogether, large PWSs provide drinking water for and thus are representative of potential exposure for approximately 77% of the US population (USEPA Safe Drinking Water Information System (SDWIS) 2017).

To update the perchlorate occurrence dataset for the 152 PWSs, two principal methods were utilized. The first method consisted of searching for and reviewing publicly available information, including consumer confidence reports (CCRs, sometimes referred to as Water Quality Reports), as well as municipal websites, and reports in the popular press. The second method consisted of contacting PWSs directly. The two methods are described in greater detail below.

METHOD 1: REVIEW OF PUBLICLY AVAILABLE INFORMATION

The Consumer Confidence Report Rule, which became effective September 18, 1998, requires all community water systems (CWSs – defined as systems serving 25 residents year-round or having 15 or more connections serving year-round residents) to provide consumers with information regarding their drinking water quality by means of an annual water quality report or CCR. Information provided to consumers includes the water source, contaminants detected in finished water (including those subject to an MCL), health effects when violations occur, likely sources of detected contaminants, and availability of source water assessments. USEPA specifies that only detected contaminants are to be included in the main water quality sampling results table of the CCR. If the PWS chooses, non-detects may be reported elsewhere in the CCR. CCRs are also to include the telephone number of the PWS

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to allow the public to obtain additional information. Other public outreach mechanisms such as public meetings also serve to fulfil public 'right-to-know' requirements of the SDWA (63 Fed Reg. 44512; USEPA 2010).

A search was conducted for the most recent available CCRs providing perchlorate data for the 72 large PWSs sampled under UCMR 1 that are located in California and Massachusetts as well as New York and Nevada. The most recent available CCR for one PWS located in New Jersey for which perchlorate concentrations were reported was also obtained. The CCRs obtained report drinking water quality from 2014 to 2016.

Internet searches were also conducted in an effort to identify publicly available information from other sources. Information was obtained for six PWSs, although some of the information duplicated information obtained from CCRs or directly from the PWSs.

METHOD 2: CONTACT PWSS DIRECTLY

Between February and September 2017 attempts were made to contact by telephone the remainder of the 152 large PWSs for which CCRs were not available (i.e., those PWSs located in states other than California, New York, Massachusetts, and Nevada). Upon establishing contact, representatives of the PWSs knowledgeable about water quality issues were asked if post-UCMR 1 perchlorate sampling took place. If post-UCMR 1 sampling took place, the PWS representatives were asked for the most recent sampling results available.

DATA QUALITY CONSIDERATIONS

Since the authors rely on data provided by others, further clarification regarding data quality is in order. As noted above, PWSs report data in CCRs in compliance with USEPA's Consumer Confidence Report Rule requiring PWSs to provide accurate information concerning drinking water quality in compliance with 'right-to-know' requirements of the SDWA. Given the reporting requirements of the SDWA, it is reasonable to infer that perchlorate data provided by PWSs to consumers by other means (e.g., PWS websites and news outlets) are also accurate. Similarly, it is also reasonable to infer that perchlorate data provided by PWSs directly to the authors are accurate. For these reasons, the data obtained were judged to be accurate at the time of reporting and reliable for the purpose of this review.

As reported by AWWA (2008), PWS distribution systems can be complex, particularly for large PWSs, with multiple sources of water, distribution system entry points, and treatment facilities. As a result, individual samples collected from specific points and times may not be fully representative of the quality of water served to all consumers at all times. However, such a limitation necessarily applies to all available perchlorate occurrence datasets, including the UCMR 1 perchlorate occurrence dataset.

The number of sample results from each PWS under UCMR 1 varied from 2 to 450 (Suffolk County Water Authority, one of the largest PWSs in the United States, is atypical of the UCMR 1 dataset), with many PWSs reporting between 4 and 8 sample results.

The number of post-UCMR 1 sample results obtained by the authors for each PWS also varied. For the purpose of this review, the authors obtained sample results from three sources: CCRs, PWS representatives, and other publicly available sources. As discussed below, sample results from CCRs constitute the majority of the updated perchlorate occurrence dataset. CCRs typically report summary statistics such as maximum and minimum concentrations. Individual sample results are not provided. A relatively small number of PWSs provided post-UCMR 1 sample results. Of the PWSs providing post-UCMR 1 sample results, the number of results varied from as few as 2 to as many as 32. In the relatively few instances in which postUCMR 1 sample results were obtained through other public sources (e.g., local news outlets), details of sampling typically were not provided.

RESULTS

Of the 152 large PWSs with perchlorate detections under UCMR 1 considered for this review, updated perchlorate data were obtained for 94 PWSs. Updated perchlorate data for 73 PWSs were obtained from CCRs and updated perchlorate data for an additional 21 PWSs were obtained via

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telephone contact and other publicly available sources of information. 44 PWSs reported that no updated perchlorate data were available. No response was received from the remaining 14 PWSs. Most of the data obtained are from the period 2014–2017, with the data obtained from CCRs limited to the period 2014–2016, but data as early as 2006 (PWSID OH2903312) are included.

Most CCRs reported a range of perchlorate concentrations based on sampling results over the preceding monitoring period. For those PWSs in which perchlorate was detected, the minimum reported perchlorate concentration typically was non-detect. For the purpose of this review, maximum reported perchlorate concentrations were conservatively used to represent updated perchlorate concentrations. For those states with MCLs (i.e., California and Massachusetts), the absence of perchlorate sample results from individual CCRs was interpreted to indicate that perchlorate was not detected, consistent with the CCR reporting requirements noted above. Perchlorate concentrations were reported in all CCRs obtained for PWSs in states without MCLs (i.e., New York, Nevada, and New Jersey).

Of the PWSs contacted by telephone, few PWS representatives were familiar with UCMR 1 sampling due to personnel changes and retirements in the intervening years since implementation of UCMR 1. However, the PWS representatives were generally familiar with current sampling practices. In response to requests for recent data, the PWSs that were able to provide perchlorate data did so in the form of laboratory reports or tabular summaries reflecting periodic (typically quarterly) monitoring activities. Most laboratory reports indicated USEPA Method 314.0 was used for analysis, although some indicated other methods were used (e.g., USEPA Method 331.0). Typically, MRLs were reported to be 4.0 μg/L, but lower MRLs were also reported, with a minimum of 0.2 μg/L. (The decrease in MRL from 4 μg/L at the time of UCMR 1 implementation to less than 1 μg/L at the present time reflects improvements in and increased experience with perchlorate analyses. Improved analytical sensitivity is consistent with the authors' experience working on perchlorate-impacted sites over the past 15 years).

In the case of 4 PWSs that were contacted, although they did not provide updated perchlorate data, they did provide contemporaneous interpretations of UCMR 1 perchlorate data, indicating that perchlorate detections were judged at the time of UCMR 1 to be unrepresentative. These PWSs emphasized considerations such as the pattern of an initial perchlorate detection followed by non-detects, the lack of an identified perchlorate source, and the inability to confirm detections using a different laboratory to support their judgment that the detections were false.

Reports of false detections in the UCMR 1 dataset obtained from PWSs is consistent with AWWA (2008). That study noted false detections for two PWSs: City of High Point, North Carolina (PWSID NC0241020) and Manatee County Utilities, Florida (PWSID FL6411132). For the City of High Point, AWWA (2008) reported that perchlorate was detected in one of eight samples collected at a concentration of 13.8 μg/L. The laboratory supervisor for the City of High Point regarded the perchlorate detection as a false positive because subsequent sampling by the City of High Point and the United States Geological Survey confirmed the absence of perchlorate in city water. Similarly, AWWA (2008) reported that the Manatee County Public Works Department attributed the detection of perchlorate in one sample to analytical errors based on the observation that perchlorate was detected only in one sample and that all other samples, which were analyzed by a different laboratory, were non-detect.

Although a direct cause-and-effect connection cannot be established based on available information, reported instances of false detections in the UCMR 1 dataset is also consistent with the tendency of USEPA Method 314.0 to produce false detections and elevated concentrations under some circumstances.

In addition to reported false detections, publicly available information identified as part of this review confirms that, in at least one instance, the initial detection of perchlorate under UCMR 1 was disregarded as unrepresentative based on the lack of subsequent detections. Specifically, the December 2004 newsletter for the City of Atlantic Beach, Florida (PWSID FL2160200) reports that samples were collected on June 28, 2004 and October 13, 2004 and that perchlorate was not detected. Review of the UCMR 1 dataset for Atlantic Beach shows the sample results reported in the December 2004 newsletter to be the last two of the 10 samples collected from the PWS under UCMR 1. Of the 10 samples, perchlorate was detected once at a concentration of 200 μg/L (August

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26, 2002). Most subsequent samples, including those on June 28, 2004 and October 13, 2004, were non-detect (City of Atlantic Beach 2004).

Aside from analytical considerations such as these, publicly available sources of information indicate that some PWS operators reported that system components (e.g., wells or water tanks) were identified to be sources of perchlorate in the PWS and removed from service. Treatment was also implemented by one PWS. The following instances were identified:

- City of Levelland, Texas (PWSID TX1100002). As reported on a local news website (KCBD 2014), an above-ground storage tank was identified as the source of perchlorate in City of Levelland drinking water and was removed from service. Upon re-testing, perchlorate was not detected.
- City of Midland, Texas (PWSID TX1650001). Detection of perchlorate in supplemental City of Midland water supply wells led to the removal of the wells from service in 2002 (Todd 2002; see also City of Midland 2017). This information was confirmed when the City of Midland was contacted and is consistent with information reported by AWWA (2008).
- City of Aberdeen, Maryland (PWSID MD0120001). In 2011, an article appearing in the Baltimore Sun, reported that the detection of perchlorate in the City of Aberdeen's PWS led to the installation of a treatment system to reduce perchlorate concentrations prior to 2009 (Wheeler 2011). The 2016 Water Quality Report for Aberdeen indicates that perchlorate concentrations have not exceeded 1 μg/L since commencement of treatment. This information was confirmed when the PWS was contacted.
- Oconee County, Georgia (PWSID GA2190000). Perchlorate was detected in water from one of six wells sampled. Subsequent to the perchlorate detection, the well in which perchlorate was detected was removed from service for reasons unrelated to the presence of perchlorate. In 2003, the Director of the Oconee County Public Utilities Department reported that, following removing the well in which perchlorate had been detected, perchlorate was no longer detected (Shearer 2003).

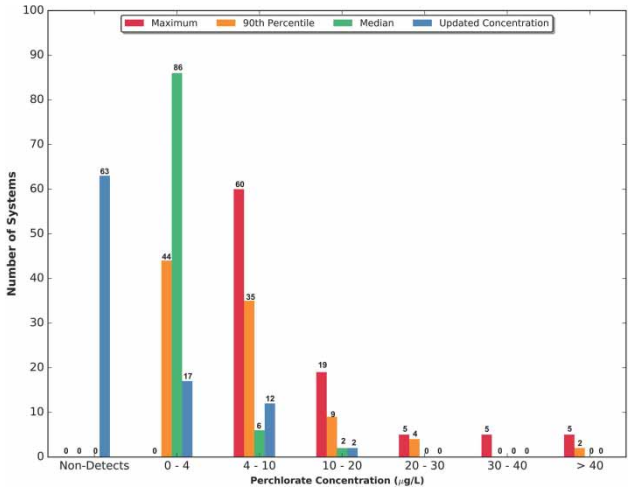

Figure 5 | Distribution of perchlorate sample results for 94 large PWSs with updated perchlorate concentrations.

Figure 5 shows the updated perchlorate concentration as well as the corresponding maximum, 90th percentile, and median concentration for each of the 94 large PWSs with perchlorate concentration updates. Figure 5 shows that of the 94 PWSs with perchlorate detections under UCMR 1, 63 PWSs are non-detect. 17 PWSs with updated perchlorate data fall in the 0–4 μg/L category, whereas UCMR 1 90th percentile and median values for 44 and 86 PWSs, respectively, fall in the 0–4 μg/L category (all maximum values from the UCMR 1 dataset are greater than 4 μg/L). Altogether, updated perchlorate concentrations are less than the maximum UCMR 1 concentrations for 90 out of the 94 PWSs. Similarly, updated perchlorate concentrations are less than the 90th percentile and median UCMR 1 concentrations for 77 and 70 out of the 94 PWSs, respectively.

Table 4 summarizes the updated perchlorate occurrence dataset by classifying the updated perchlorate concentrations using the same concentration categories used in Table 3 (with the addition of the non-detect category). Table 4 also summarizes the 2017 US population associated with the PWSs in each concentration category. To allow direct comparison of the updated perchlorate occurrence dataset with the UCMR 1 dataset, Table 4 lists perchlorate concentrations (maximum, 90th percentile, and median) from the original UCMR 1 dataset for the same 94 large PWSs for which updates are available. Table 4 shows that

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Table 4 | Population served By large United States public water systems with at aeast one perchlorate detection under UCMR 1 – updated maximum concentrations

	Population served by PWS with updated concentrations (Total population = 12,668,000)							
	Maximum[a]		90th Percentile[a]		Median[a]		Updated concentration	
Perchlorate threshold (μg/L)	Population	Percentage PWSs with at least 1 detection greater than threshold	Population	Percentage PWSs with at least 1 detection greater than threshold	Population	Percentage PWSs with at least 1 detection greater than threshold	Population	Percentage PWSs with at least 1 detection greater than threshold
Non-detect	0	0% (0 out of 94)	0	0% (0 out of 94)	0	0% (0 out of 94)	8,829,000	70
0–4	0	0.0	8,249,000	65	11,813,000	93	1,448,000	12
4–10	7,856,000	62	2,951,000	23	547,000	4	2,329,000	18
10–20	3,135,000	25	361,000	2.8	307,000	2.4	61,000	0.5
20–30	518,000	4.1	1,049,000	8.3	0	0	0	0
30–40	704,000	5.6	0	0	0	0	0	0
>40	454,000	3.6	58,000	0.5	0	0	0	0

Notes:
1. Non-detects were set to half the detection limit, 2 μg/L.
2. Population numbers have been rounded to the nearest thousand.
3. Source of population data: EPA SDWIS Federal Reports.
4. Population data from the Metropolitan Water District of Southern California (MWD) were excluded from the above table to avoid double counting. MWD supplies water to many PWSs in Southern California, including some PWSs under consideration here.
5. Most data are from 2014–2017, with small amount of data from as early as 2006.
[a]UCMR 1 results for those PWSs with updated perchlorate concentrations.
PWSs – Public Water Systems.
UCMR 1 – Unregulated Contaminant Monitoring Rule 1.

approximately 12.7 million people were served by the 94 large PWSs with perchlorate detections under UCMR 1 (i.e., were potentially exposed to perchlorate). The updated perchlorate dataset shows that approximately 8.8 million are no longer potentially exposed to perchlorate, representing a 70% reduction in potential exposure. Table 4 also shows that the potential exposure of the US population provided water by the 94 large PWSs with perchlorate detections has generally decreased in each concentration category using maximum, 90th percentile, and median UCMR 1 perchlorate concentrations.

Figure 6 shows the updated distribution of perchlorate occurrence across the USA, Puerto Rico, and the Northern Marianas Islands for those PWSs for which updated perchlorate data are available. Comparison of Figure 6 with Figures 2–4 shows that, for those 94 large PWSs with updated perchlorate data, the geographic extent of perchlorate detections using the updated perchlorate dataset is less than that using the maximum, 90th percentile, and median concentrations from the UCMR 1 dataset.

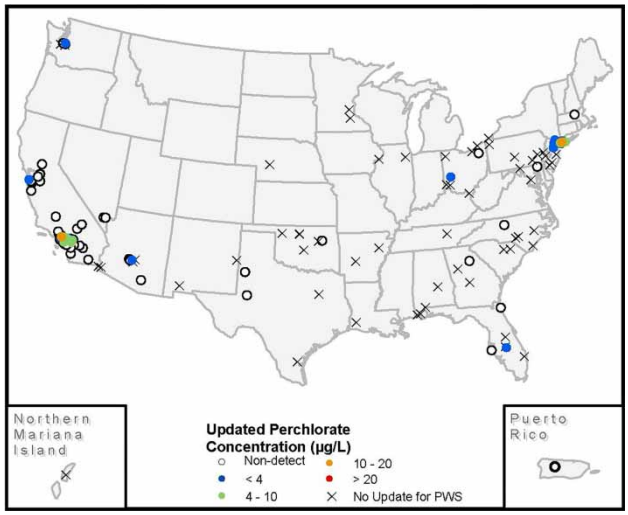

Figure 6 | Updated perchlorate detections for large PWSs.

DISCUSSION

This review has resulted in the development of an updated perchlorate occurrence dataset for 94 of the 152 large

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PWSs with perchlorate detections under UCMR 1. Comparison of the updated and UCMR 1 perchlorate datasets shows that perchlorate concentrations have changed since implementation of UCMR 1 and that the UCMR 1 dataset is no longer representative for those PWSs for which more recent data are available. Comparison of the updated and UCMR 1 perchlorate datasets also shows that the following changes have taken place since implementation of UCMR 1:

- Perchlorate concentrations in most large PWSs with perchlorate detections under UCMR 1 have decreased.
- The US population potentially exposed to perchlorate in large PWSs with perchlorate detections under UCMR 1 has decreased.
- The geographic distribution of perchlorate occurrence for those large PWSs with perchlorate detections under UCMR 1 has decreased.

The reductions in perchlorate occurrence since UCMR 1 suggest that regulation of perchlorate by USEPA may not be warranted at this time.

There are a number of sources of uncertainty in the interpretation and comparison of the UCMR 1 and updated perchlorate datasets. The largest source of uncertainty in the updated perchlorate occurrence dataset appears to be the relatively limited number of updated sample results available for some PWSs (i.e., some results reported by news outlets). However, CCRs, which provided approximately 75% (73 out of 94 PWSs) of updated perchlorate occurrence data, are relatively robust datasets of sufficient quality to meet SDWA reporting requirements. Therefore, uncertainties associated with data limitations seem relatively minor, particularly in light of the relatively large differences between UCMR 1 and updated perchlorate datasets.

Uncertainties related to analytical methods could also impact the results of this review. However, given improvements in analytical methods and the relatively large number of non-detects in the updated perchlorate occurrence dataset, it seems unlikely that analytical issues such as false detections are significant.

Moreover, the authors sought to reduce the potential impacts of uncertainties on the results of this review by conservatively selecting maximum reported perchlorate concentrations to represent the updated perchlorate concentration for each PWS. Selection of maximum concentrations for the updated perchlorate occurrence dataset would tend to reduce differences between the UCMR 1 and updated perchlorate occurrence datasets. Such conservatism notwithstanding, the differences between UCMR 1 and updated perchlorate datasets is relatively large.

Changes that appear to have resulted in the decrease of perchlorate occurrence appear to include the following: state-level regulation of perchlorate in drinking water (i.e., establishment of MCLs in California and Massachusetts), as well as remediation of perchlorate impacts, and/or provision of treated or alternative water supplies (e.g., the City of Aberdeen) have resulted in a reduction in perchlorate occurrence in drinking water in the United States. In addition, improved analytical methods (i.e., USEPA Methods 314.1 and 331.0) as well as greater experience with older methods (i.e., USEPA Method 314.0) have likely resulted in more accurate assessment of perchlorate occurrence and a reduction in the incidence of false or elevated detections since the time of UCMR 1 implementation.

The European Union is another jurisdiction that has evaluated perchlorate occurrence. In 2014, the European Food Safety Authority (EFSA) issued a scientific opinion regarding the potential risks to public health related to the presence of perchlorate in food. In its opinion, the EFSA concluded that perchlorate exposures were of potential concern. However, occurrence data were relatively limited. As a consequence, the European Commission's Directorate-General for Health and Food Safety indicated a need for additional data in 2015. Meanwhile, in recognition of the desire to promote trade among European Union Member States, provisional perchlorate enforcement levels in broad food categories were also provided for use concurrently with continued sample collection. In 2017, the EFSA completed a more extensive assessment of potential human exposure to perchlorate, utilizing results of samples collected after September 1, 2013 throughout the European Union (EFSA CONTAM 2014; European Commission Directorate-General for Health and Food Safety 2015; Arcella *et al.* 2017).

Thus, consistent with the United States, regulatory decision-making in Europe can be a multi-year process. So far, however, the European decision-making process spans only a few years. It remains to be seen if the European Union will arrive at a final perchlorate determination before conditions change.

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CONCLUSIONS

As this review has also shown for the case of perchlorate in drinking water in the United States, regulatory rule-making tends to be a stepwise, deliberate process requiring years to complete. Given the complexities of regulatory rule-making, associated costs of implementation, and, of course, potential impacts on public health, thoughtful, deliberate regulatory decision-making is appropriate. However, the very conditions that prompted consideration of new regulations can change while those new regulations are being considered. In the case of perchlorate in the United States, conditions improved. In other circumstances, of course, conditions could worsen.

As this review has also shown, not only can conditions in the natural world change, but conditions in institutions change as well. Personnel changes and retirements, for example, result in loss of institutional knowledge. Tabulated data summaries may be saved, but laboratory reports, field notes, and related information typically are not preserved. As a consequence, gathering and interpreting data becomes more challenging over time.

To generalize, the results of this review of perchlorate occurrence in the United States suggest that as part of a long-term regulatory decision-making process, regulatory bodies should periodically re-evaluate the conditions that prompted initiation of the decision-making process. The use of provisional regulations in parallel with data-gathering efforts, as in the European Union example, provides an approach that may increase the likelihood that regulations will remain 'on target' and appropriately address conditions of concern.

Since the rate at which conditions change varies, identification of a general threshold triggering such re-evaluations is not feasible. However, the review of perchlorate occurrence in drinking water in the United States presented in this paper suggests that after the passage of 10 years or more a re-evaluation is warranted.

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