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Sociodemographic inequalities in uranium and other metals in community water systems across the USA, 2006–11: a cross-sectional study

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Summary

Background—The US Environmental Protection Agency (EPA) currently sets maximum contaminant levels (MCLs) for ten metals or metalloids in public drinking water systems. Our objective was to estimate metal concentrations in community water systems (CWSs) across the

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Contributors

FR, BCB, SNC, KS, AB, AN-A, and AEN conceived the study. FR, AN-A, and AEN developed the methodology. BCB, SNC, KS, and AB contributed to developing the methodology. FR conducted the investigation (performed data analysis). AEN led the investigation (developed and performed the data analysis). AN-A led the investigation (developed the data analysis approach). FR, YY, and AEN managed data and formally analysed data. FR, YY, BCB, SNC, KS, AB, AN-A, and AEN contributed to manuscript writing. YY developed the interactive website and software. AN-A and AEN supervised the study. FR and AEN accessed and verified the underlying data. All authors had access to the data and had final responsibility for the decision to submit for publication.

For the Spanish translation of the abstract see **Online** for appendix 1

For the **EPA National Primary Drinking Water Regulations** see https://www.epa.gov/ground-water-and-drinking-water/national-primary-drinking-water-regulations

For the **EPA webpage on public water systems** see https://www.epa.gov/dwreginfo/information-about-public-water-systems

For the **SYR2 compliance monitoring data** see https://www.epa.gov/dwsixyearreview/six-year-review-2-contaminantoccurrence-data-1998-2005

For the **SYR3 compliance monitoring data** see https://www.epa.gov/dwsixyearreview/six-year-review-3-compliance-monitoring-data-2006-2011

See **Online** for appendix 2

For the **SDWIS Federal Reporting Services system** see https://www.epa.gov/ground-water-and-drinking-water/safe-drinking-water-information-system-sdwis-federal-reporting

For the **interactive map and dashboard** see https://msph.shinyapps.io/drinking-water-dashboard/

For the **online dashboard** see msph.shinyapps.io/drinking-water-dashboard/

For **datasets of the average metal concentrations** see https://github.com/annenigra/US-PublicWaterSystem-Metal-Estimates

See **Online** for appendix 3

Declaration of interests

We declare no competing interests.

USA, to establish if sociodemographic or regional inequalities in the metal concentrations exist, and to identify patterns of concentrations for these metals as a mixture.

Methods—We evaluated routine compliance monitoring records for antimony, arsenic, barium, beryllium, cadmium, chromium, mercury, selenium, thallium, and uranium, collected from 2006–11 (2000–11 for uranium; timeframe based on compliance monitoring requirements) by the US EPA in support of their second and third Six-Year Reviews for CWSs. Arsenic, barium, chromium, selenium, and uranium (detectable in >10% records) were included in the main analyses (subgroup and metal mixture analyses; arsenic data reported previously). We compared the mean, 75th percentile, and 95th percentile contaminant concentrations and the percentage of CWSs with concentrations exceeding the MCL across subgroups (region, sociodemographic county-cluster, size of population served, source water type, and CWSs exclusively serving correctional facilities). We evaluated patterns in CWS metal concentration estimate profiles via hierarchical cluster analysis. We created an online interactive map and dashboard of estimated CWS metal concentrations for use in future analyses.

Findings—Average metal concentrations were available for a total of 37 915 CWSs across the USA. The total number of monitoring records available was approximately 297 000 for arsenic, 165 000 for barium, 167 000 for chromium, 165 000 for selenium, and 128 000 for uranium. The percentage of analysed CWSs with average concentrations exceeding the MCL was 2·6% for arsenic (MCL=10 μg/L; nationwide mean 1·77 μg/L; n=36 798 CWSs), 2·1% for uranium (MCL=30 μg/L; nationwide mean 4·37 μg/L; n=14 503 CWSs), and less than 0·1% for the other metals. The number of records with detections was highest for uranium (63·1%). 75th and 95th percentile concentrations for uranium, chromium, barium, and selenium were highest for CWSs serving Semi-Urban, Hispanic communities, CWSs reliant on groundwater, and CWSs in the Central Midwest. Hierarchical cluster analysis revealed two distinct clusters: an arsenic–uranium–selenium cluster and a barium–chromium cluster.

Interpretations—Uranium is an under-recognised contaminant in CWSs. Metal concentrations (including uranium) are elevated in CWSs serving Semi-Urban, Hispanic communities independent of location or region, highlighting environmental justice concerns.

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Introduction

Chronic exposure to metals, including uranium, is associated with several adverse health outcomes including liver damage, nephrotoxicity, and cardiovascular disease.[1–5] In the USA, the Environmental Protection Agency (EPA) sets maximum contaminant levels (MCLs) for six classes of contaminants, including ten metals or metalloids, in public drinking water systems in accordance with the EPA Safe Drinking Water Act (SDWA). However, nationwide estimates of metal concentrations in public drinking water systems are only available for arsenic.[6,7] Most US residents rely on public drinking water systems, with most residents (approximately 90%) relying specifically on community water systems (CWSs) that serve the same population year round.[8] Violations of EPA SDWA regulations (eg, MCL exceedances or inadequate public notification of violations) are relatively common. One

study reported that more than half of all CWSs reported an SDWA violation during a 1-year period (fiscal year 2011).[9,10]

Racial and socioeconomic disparities in drinking water access and quality are related to structural inequalities in the built environment, land use and planning policies, and differences in the geological environment (eg, racial or ethnic subgroups are not uniformly distributed across the USA, therefore some geological conditions disproportionately affect particular populations).[11,12] Inequalities in concentrations of regulated contaminants in public water systems across racial or ethnic and socioeconomic subgroups of the US population have been described in detail for arsenic and nitrates.[6,13] Hispanic communities, tribal communities, and communities in the southwestern USA are more likely to be served by CWSs that exceed arsenic and nitrate MCLs.[6,13] CWSs reliant on groundwater that serve small communities are also more likely to exceed the arsenic MCL.[6] Systematic nationwide studies of potential inequalities in public drinking water contaminant concentrations across population subgroups have not been conducted for other regulated metal contaminants. Examining potential spatial and demo graphic disparities in public drinking water contaminant concentrations can inform public health interventions and regulatory actions to reduce exposure inequalities, and can possibly identify relevant exposure sources that might contribute to inequalities in overall metal exposures and related adverse health outcomes.

The objectives of this study were to estimate CWS metal concentrations across the USA; identify sociodemographic subgroups served by CWSs that either reported high metal concentration estimates or were more likely to report averages exceeding an MCL; and characterise metal mixture profiles in CWSs nationwide. We examined antimony, arsenic, barium, beryllium, cadmium, chromium, mercury, selenium, thallium, and uranium. Because the concentrations of antimony, beryllium, cadmium, mercury, and thallium were low and rarely exceeded MCLs (less than 10% of records were detectable for these metals), we focused our analysis on arsenic, barium, chromium, selenium, and uranium. We estimated metal concentrations at the CWS level using the compliance monitoring data supporting EPA's second (2000–05) and third (2006–11) Six-Year Reviews (SYR2 and SYR3) of drinking water regulations, which contain routine compliance monitoring records for public water systems. We focused on CWSs that serve most of the US population year round. Because previously published analyses of CWS arsenic concentrations identified inequalities across US region, sociodemographic county clusters, population-served size, source water type, and CWSs which exclusively serve correctional facilities,[6,7] we examined these same subgroups in our analysis.

Methods

Data source

We used CWS routine compliance monitoring records published in the US EPA's database supporting the SYR2 and SYR3 to estimate CWS metal concentrations, following a previously published protocol.[6] Details regarding the SYR databases and the development of CWS-level metal concentrations are available in appendix 2 (pp 2–4). Monitoring data from the SYR3 period (2006–11) includes approximately 13 million analytical records from 139 000 public water systems serving 290 million people annually. Records represent

95% of all public water systems and 92% of the total population served by public water systems nationally.[14–16] We used the SYR3 records to develop CWS estimates for all metals, and additionally included records from SYR2 (2000–05) to develop estimates for uranium (compliance monitoring requirements for uranium are different than those for other metals because the MCL for uranium [30 μg/L] was established in 2000 under the EPA Radionuclides Final Rule; appendix 2 p 2).[14,17,18]

CWS-level metal concentration estimates

Metal concentrations lower than the record-specific limit of detection (LOD) were replaced by the LOD divided by the square root of two (this method is used by the US Centers for Disease Control and Prevention and other federal agencies when reporting geometric or arithmetic means of environmental biomarkers and concentrations; appendix 2 p 3).[19] The percentage of records with values higher than the LOD was 2·2% for antimony, 45·5% for arsenic, 60·8% for barium, 1·3% for beryllium, 1·6% for cadmium, 18·9% for chromium, 1·5% for mercury, 12·9% for selenium, 1·6% for thallium, and 63·1% for uranium. We restricted our main analyses (subgroup analyses and metal mixture analyses) to five metals (arsenic, barium, chromium, selenium, and uranium) with more than 10% of records above the LOD.

For each metal, many CWSs reported multiple monitoring records per year. We first calculated mean CWS metal concentrations within each calendar year. When the mean concentration of metals in treated water samples was lower than in untreated samples, we calculated the annual mean with treated samples only (few CWSs reported records for both treated and untreated samples within the same year). Because uranium records from SYR2 did not distinguish between treated and untreated samples, uranium estimates only accounted for treatment in records from 2006–11. We then averaged CWS metal concentrations to 2006–11 (SYR3 period). For uranium, we averaged concentrations to 2000–11, which covers grandfathered or initial compliance samples (2000–07) and samples collected during the first compliance monitoring cycle (2008–16). We compared findings from several sensitivity analyses that aggregated metal concentrations to different periods, all with similar results (appendix 2 pp 4, 8).

We then merged CWS metal concentrations with system inventory information extracted from the EPA Safe Drinking Water Information System (SDWIS), including counties served, number of people served, and source water type, as previously described in detail.[6]

Nationwide analysis

All data management and analysis was conducted in R (version 3.5.3). We calculated the distribution, including percentiles and arithmetic means, of 6-year (2006–11) average water concentrations for each metal (2000–11 for uranium) at the CWS level for the entire USA. Our analysis focuses on evaluating 75th and 95th percentile values because, firstly, median concentration values were below the LOD for these metals; secondly, arithmetic mean concentrations are influenced by the high number of records at or below the LOD, whereas higher percentile values are not; and thirdly, measures of central tendency do not reflect percentiles at the highest end of the distribution that affect the most exposed

populations and are particularly relevant for population-level environmental exposures.[6,20] We also calculated the number and percentage of CWSs with concentrations exceeding the WHO *Guidelines for Drinking-water Quality* limit or the EPA MCL for each metal.[21]

Stratification and analysis by subgroups

To identify subgroups of the US population whose estimated CWS metal concentrations were relatively high, we also calculated the 75th percentile, 95th percentile, and mean concentration and 95% CI for each metal in analyses stratified by select population subgroups: source water type (groundwater *vs* surface water as reported in SDWIS; only 1% of systems reported to use both, which were categorised as groundwater); size of the population served (standard EPA categories ≤500 people, 501–3300 people, 3301–10 000 people, 10 001–100 000 people, and >100 000 people); CWSs exclusively serving correctional facilities (identified via a keyword search used previously[7]); region (Pacific Northwest, Southwest, Central Midwest, Eastern Midwest, Southeast, Mid-Atlantic, New England, and Alaska and Hawaii; groupings were based on a previous analysis for arsenic[22]); and sociodemographic county-cluster. Sociodemographic county-clusters (n=8 clusters) were derived by Wallace and colleagues[23] to enable the direct comparison of county-level health outcomes while accounting for the sociodemographic makeup of a county's population (eg, racial and ethnic composition, urbanicity, insurance coverage, age), and have been used in a previous analysis of CWS arsenic inequalities.[6] We stratified CWS metal concentration estimates by these sociodemographic county-clusters to identify characteristics of broad population subgroups exposed to elevated CWS metal concentrations. The sociodemographic county-clusters are: Semi-Urban, High Socioeconomic Status (SES); Semi-Urban, Middle-to-Low SES; Semi-Urban, Hispanic; Mostly Rural, Middle SES; Rural, Middle-to-Low SES; Young, Urban, Middle-to-High SES; Rural, American Indian; and Rural, High SES. We also plotted the distribution of mean CWS uranium concentrations across regions to evaluate if distribution shapes were similar.

We assessed whether CWS metal distributions were significantly different across subgroups via non-parametric Kruskal-Wallis tests (all CWS metal distributions were log-normal). Because our initial analysis identified the highest uranium concentrations in CWSs serving Semi-Urban, Hispanic counties and CWSs located in the Southwest, we conducted several post-hoc analyses to identify if increased uranium concentrations for CWSs serving Semi-Urban, Hispanic counties could be explained by geography and geology. We compared the 75th, 90th, and 95th percentiles and arithmetic mean uranium concentrations for CWSs serving Semi-Urban, Hispanic counties versus CWSs serving all other types of counties separately within the states of California, Texas, and Oklahoma, as these were the states with the highest CWS uranium concentrations of all states located in the Southwest. We also evaluated the change in 90th percentile and arithmetic mean CWS uranium concentration (dependent variable) per 1% higher proportion of the county population classified as Hispanic/Latino (independent variable) using 2010 US Census Bureau statistics,[24,25] adjusting for state (categorical), the size of the population served (continuous), and the source water type (surface *vs* groundwater) via quantile regression using the quantreg package (version 5.85) in R. Quantile regression quantifies associations that occur in the tails of the distribution, and is commonly used in analyses of environmental exposures,

which often have skewed distributions.[20] This analysis was conducted for all US counties, for counties classified as Semi-Urban, Hispanic, and for counties located in the Southwest.

County-level maps

To visually identify spatial patterns in metal concentration estimates across the USA, we also estimated county-level, population-weighted CWS metal concentrations, as previously described in detail, with county-level concentrations weighted by the number of people served by each CWS within a county (appendix 2 pp 5–6).[6] Because only the county served was reliably reported in SDWIS for each CWS, we could not aggregate to smaller geographic scales (eg, census tract). We mapped county-level estimates of 6-year (2006–11) average water concentrations for each metal (2000–11 for uranium) across the conterminous USA using the maps package (version 3.3.0) in R.

We created an interactive map and dashboard of estimated metal concentrations at the CWS and county levels for use in future analyses.

Hierarchical clustering

We conducted analyses to evaluate the composition of metals in CWSs as a complex mixture. We first calculated Spearman's correlation coefficients between all metal pairs after log transformation. Subsequent analysis was restricted to CWSs with concentration estimates available for all five main metals of interest. To identify distinct homogenous subgroups of metals, we conducted hierarchical cluster analysis to combine the metals into agglomerative clusters.[26] We used Ward's method for Euclidean distances and normalised each metal to unit variance and zero mean before constructing dendrograms to assess the cohesiveness of the cluster using the R package ggdendro (version 0.1.22). Because CWSs were most likely to be missing uranium concentrations (SYR3 only covers years 2006–11 and the First Radionuclides Rule Compliance Cycle covers 2008–16), we conducted a sensitivity analysis repeating our hierarchical cluster analysis for CWSs with concentration estimates for the other four metals (n=34 284 CWSs).

Role of the funding source

The funders of the study had no role in study design, data collection, data analysis, data interpretation, or writing of the report.

Results

Average metal concentrations were available for 37 915 CWSs across the USA. For each of the ten metals, the total number of records ranged from approximately 128 000 (uranium) to 297 000 (arsenic; table 1). Of all the metals examined, the number of records with detections was highest for uranium (63·1%). Arsenic had the largest proportion of average CWS concentrations above the US EPA MCL (2·6%), followed by uranium (2·1%).[6,7] All other metals examined had less than 0·1% of average CWS concentrations above the MCL. We describe the five metals included in our main analyses (detectable in >10% records) herein. Nation wide, the 75th (and 95th) percentile of metal concentrations from 2006–11 was 1·66 μg/L (7·40 μg/L) for arsenic, 79 μg/L (253 μg/L) for barium, 0·62 μg/L (5·30 μg/L)

for chromium, less than 0·60 μg/L (3·81 μg/L) for selenium, and 3·1 μg/L (18·5 μg/L) for uranium (2000–11). Subgroup-specific results and county-level maps for arsenic have been described in previous publications, but are included in tables and figures for comparison.[6,7]

Metal concentration estimates were stratified by source water type, CWS size, and correctional facilities. CWSs reliant on groundwater had higher 75th and 95th percentile concentrations than CWSs reliant on surface water for barium (75th percentile, 87 μg/L *vs* 40 μg/L; 95th percentile, 265 μg/L *vs* 107 μg/L), chromium (0·70 μg/L *vs* 0·27 μg/L; 5·59 μg/L *vs* 2·36 μg/L), selenium (<0·60 μg/L *vs* <0·60 μg/L; 4·10 μg/L *vs* 2·10 μg/L), and uranium (3·4 μg/L *vs* 1·5 μg/L; 19·5 μg/L *vs* 7·1 μg/L), although differences were not statistically significant for selenium (table 2). Compared with CWSs serving larger populations, those serving smaller populations generally had higher concentrations of arsenic, barium, and uranium; whereas, these patterns were more mixed for chromium and selenium. CWSs serving up to 500 people had the highest 75th and 95th percentile concentrations of arsenic (75th percentile 1·90 μg/L, 95th percentile 8·08 μg/L) and uranium (3·5 μg/L, 20·7 μg/L), and CWSs serving 501–3300 people had the highest 75th and 95th percentile concentrations of barium (94 μg/L, 275 μg/L). Results were similar when comparing the arithmetic mean (appendix 2 p 10).

Nationwide, distributions of metal concentrations for CWSs exclusively serving correctional facilities were similar to those for all CWSs for arsenic, barium, selenium, and uranium ($p>0·05$), but were significantly different for chromium (table 2). CWSs serving correctional facilities had higher 75th and 95th percentile concentrations of chromium (1·33 μg/L, 7·04 μg/L) than those for all CWSs (0·62 μg/L, 5·30 μg/L; $p=0·042$). When comparing the arithmetic mean, CWS concentration estimates for correctional facility CWSs were significantly different for barium and uranium, but not chromium (appendix 2 p 10).

Metal concentration estimates were stratified by region and sociodemographic county cluster. CWS concentration estimates (75th and 95th percentiles) were highest in the Central Midwest region for all metals except arsenic and barium (table 2). 75th and 95th percentile concentrations for barium in the Central Midwest were 149 μg/L and 304 μg/L, with the next highest (75th percentile) concentrations in the Southwest (115 μg/L, 270 μg/L) and the Eastern Midwest (111 μg/L, 309 μg/L). 75th and 95th percentile concentrations for chromium in the Central Midwest were 3·10 μg/L and 8·69 μg/L, with the next highest concentrations in the Southwest (1·49 μg/L, 7·68 μg/L) and Alaska and Hawaii (1·10 μg/L, 3·63 μg/L). 75th and 95th percentile concentrations for selenium in the Central Midwest were 2·71 μg/L and 12·50 μg/L, with the next highest concentrations in the Southwest (1·03 μg/L, 6·50 μg/L) and the Eastern Midwest (1·01 μg/L, 3·54 μg/L). 75th and 95th percentile concentrations for uranium in the Central Midwest were 11·4 μg/L and 32·2 μg/L, with the next highest concentrations in the Southwest (9·6 μg/L, 28·3 μg/L) and New England (4·0 μg/L, 25·1 μg/L). When comparing the arithmetic mean across regions, some differences in regional ordering were observed for each metal (eg, mean uranium concentrations were highest in the Southwest followed by the Central Midwest; appendix 2 p 10). The distribution of average uranium concentrations was right skewed for all regions except the Central Midwest (bimodal) and the Southwest (relatively uniform; appendix 2 pp 13–14). At the county-level, all four metals showed a general pattern of higher concentrations in central

and western counties versus eastern counties (figure 1). Because the SYR3 only covers years 2006–11 and the First Radionuclides Rule Compliance Cycle covers 2008–16, uranium has the highest proportion of missing data, which is reflected in the relatively poor spatial coverage for the Eastern Midwest, Southeast, Mid-Atlantic, and New England regions.

CWSs serving Semi-Urban, Hispanic counties had the highest 75th and 95th percentile concentrations for all metals (table 2). 75th and 95th percentile barium concentrations for CWSs serving Semi-Urban, Hispanic counties were 110 μg/L and 279 μg/L, with the next highest concentrations in CWSs serving Young, Urban, Middle-to-High SES counties (110 μg/L, 227 μg/L) and Rural, High SES counties (100 μg/L, 266 μg/L). 75th and 95th percentile chromium concentrations for CWSs serving Semi-Urban, Hispanic counties were 1·49 μg/L and 8·00 μg/L, with the next highest concentrations in CWSs serving Rural, High SES counties (1·38 μg/L, 6·35 μg/L) and Rural, American Indian counties (0·98 μg/L, 5·35 μg/L). 75th and 95th percentile selenium concentrations for CWSs serving Semi-Urban, Hispanic counties were 1·27 μg/L and 8·49 μg/L, with the next highest concentrations in CWSs serving Rural, High SES counties (0·82 μg/L, 7·21 μg/L) and Rural, American Indian counties (0·74 μg/L, 4·85 μg/L). 75th and 95th percentile uranium concentrations for CWSs serving Semi-Urban, Hispanic counties were 10·9 μg/L and 31·7 μg/L, with the next highest concentrations in CWSs serving Young, Urban, Middle-to-High SES counties (6·3 μg/L, 17·7 μg/L) and Rural, High SES counties (3·7 μg/L, 22·8 μg/L). Comparing the arithmetic mean across these sociodemographic clusters produced different rankings across the clusters; however, CWSs serving Semi-Urban, Hispanic counties also had the highest arithmetic mean concentration for all metals (appendix 2 pp 10–11).

We did a post-hoc analysis of uranium concentrations in CWSs serving Semi-Urban, Hispanic counties versus CWSs serving all other counties in three Southwest states (California, Oklahoma, and Texas). The 75th and 95th percentile uranium concentrations were higher among the CWSs serving Semi-Urban, Hispanic counties in California (11·7 μg/L, 35·1 μg/L *vs* all other CWSs, 6·4 μg/L, 18·3 μg/L), Oklahoma (11·4 μg/L, 49·0 μg/L *vs* all other CWSs, 1·9 μg/L, 11·3 μg/L), and Texas (20·6 μg/L 44·6 μg/L *vs* all other CWSs, 10·5 μg/L, 24·5 μg/L; table 3). We also conducted a post-hoc quantile regression analysis. Per 1% higher proportion of the county population classified as Hispanic/Latino, 90th percentile uranium concentration increased by 15·1 μg/L among all CWSs ($p<0·0001$), by 25·9 μg/L among CWSs in the Southwest ($p<0·0001$), and by 11·2 μg/L among CWSs serving Semi-Urban, Hispanic counties ($p=0·22$; table 4). Results were similar when assessing arithmetic means (appendix 2 p 12).

Regarding metal mixtures in CWSs nationwide, we observed moderately positive Spearman's correlations between arsenic and selenium ($r=0·33$), arsenic and uranium ($r=0·25$), and chromium and selenium ($r=0·33$; appendix 2 p 15). Hierarchical cluster analysis of CWSs with concentration estimates available for the five metals of interest (n=12 756 CWSs) revealed two distinct clusters: arsenic–selenium–uranium and barium–chromium (figure 2). Sensitivity analyses repeating the hierarchical cluster analysis without uranium (n=34 284 CWSs) yielded a similar arsenic–selenium cluster and a barium–chromium cluster (not shown).

Discussion

The current study indicates that although most regulated metals are rarely measured in US CWSs above detection limits and MCLs (especially antimony, beryllium, cadmium, mercury, and thallium), substantial geographic and sociodemographic variability exists for CWS uranium concentrations. We estimated that 63·1% of CWS compliance monitoring records reported detectable concentrations of uranium, and that 2·1% of CWSs with available uranium data had 2000–11 average concentrations above the MCL (smaller than the percentage of wells—approximately 4%—exceeding the uranium MCL in the US National Water Information System).[27] Despite relatively frequent detections and relatively high concentrations compared with other metals in our study (highest arithmetic mean, 4·37 μg/L), uranium has been underappreciated in the literature as a public drinking water contaminant of concern.

Consistent with previous findings for arsenic,[6,7] CWSs reliant on groundwater had higher mean and 95th percentile concentrations for barium, chromium, selenium, and uranium compared with CWSs reliant on surface water. Mean and 95th percentile concentrations were also higher for CWSs serving smaller populations compared with those serving the largest populations for arsenic, barium, and uranium. CWSs serving smaller populations are likely to have few financial and technical resources available to implement aggressive treatment techniques, or source water switching or mixing for many types of regulated contaminants.[6,10,28] Additionally, some treatment techniques and source water changes implemented in accordance with MCL changes (eg, arsenic in 2006 and uranium in 2008) might have reduced or influenced the concentration of other metals in CWSs. Future analyses could evaluate whether CWSs that greatly reduced arsenic concentrations in accordance with the 2006 MCL change subsequently report reduced concentrations of other metals.

The current analysis also revealed significant spatial variability and inequalities in CWS uranium concentrations, which probably reflects local geological context. Although most regulated metals are relatively geologically rare and are associated with specific environments not widely disseminated throughout the USA, uranium, selenium, and arsenic are all relatively common at measurable concentrations in widely disseminated conditions. The release of uranium, selenium, and arsenic in groundwater is dependent on the redox environment, which controls both spatial distribution and temporal evolution of groundwater metal concentrations. Both uranium and selenium are highly soluble as oxidised species in groundwater (U[VI] and Se[VI]/Se[IV]), while the reduced species (U[IV] and Se[0]/Se[–II]] are insoluble at near-neutral pH. Thus, uranium and co-occurring selenium with similar redox potential are mobilised by oxidative dissolution encountered in oxic groundwater. In contrast, reducing conditions immobilise uranium and selenium and often lead to release of arsenic by reductive dissolution of iron (Fe[III]) oxyhydroxides.[29,30] This contrast in mobilisation conditions suggests that arsenic contamination is most common under reducing conditions, while uranium and selenium are most soluble under oxidising conditions.

Our hierarchical cluster analysis revealed the presence of a strong uranium–selenium–arsenic cluster and a barium–chromium cluster, possibly related to redox conditions. Barium

is not redox active and most barium salts are insoluble, whereas chromium is typically found primarily as insoluble Cr(III) under almost all anoxic groundwater conditions.[31] Arsenic and uranium can both be elevated in groundwater samples,[32–34] and previous studies have found that arsenic and uranium co-occur in unregulated drinking water on Navajo Nation[29,35] and in untreated public supply wells across the USA,[36] potentially pointing to the importance of carbonato complexes in particular in increasing solubility. This solubility effect is much better understood for uranium than arsenic due to extensive modelling of uranium transport,[37–39] and has been observed[40] but insufficiently described for arsenic transport. In many cases, aqueous uranium and arsenic are also found in forms such as carbonate complexes[40,41] that do not strongly adsorb to iron or aluminium oxides, which is the most commonly used water treatment method to remove chemical contaminants. Thus, the presence of bicarbonate ions in the oxic ground and surface water sources could also explain the persistence of arsenic and uranium in CWSs that we observed despite treatment.

Our findings for CWSs serving Semi-Urban, Hispanic communities further highlight the substantial environmental justice concerns for Hispanic/Latino communities raised in previous studies of CWS arsenic and nitrate concentrations.[6,7,13] Although geological variability might explain much of the regional differences in nationwide uranium spatial patterns, it does not account for disparities across sociodemographic county-clusters. Compared with CWSs serving other sociodemographic groups, CWSs serving Semi-Urban, Hispanic communities had the highest uranium, selenium, barium, chromium, and arsenic concentrations. Furthermore, quantile regression analyses indicated a significant increase in 90th percentile and mean CWS uranium concentrations per 1% higher proportion of the population classified as Hispanic/Latino for all CWSs, and for CWSs in the Southwest, after adjusting for state, size of the population served, and source water type. These findings indicate that inequalities in CWS uranium concentrations for Hispanic/Latino communities are not merely due to geographic location, groundwater use, or CWS size. Although the chemistry of these metals vary widely and they originate from a variety of sources, the consistent association between elevated CWS metal concentrations and Semi-Urban, Hispanic communities implies that concentration disparities are a failure of regulatory policy or treatment rather than underlying geology. Hispanic/Latino populations show numerous health disparities including increased mortality due to diabetes, liver disease, and kidney disease.[42] Hispanic/Latino populations have lower all-cause, cardiovascular, and cancer mortality rates than US non-Hispanic White populations despite overall poorer health-care access and lower socioeconomic status, which is sometimes referred to as the Hispanic paradox. Hispanic/Latino communities are incredibly diverse by national origin, dietary patterns, language, and other relevant social and environmental determinants of health.[43] Future analyses should explore CWS differences within Hispanic/Latino communities, whether disproportionate chronic, low-level CWS metal exposure contributes to inequalities in associated adverse health outcomes, and whether these CWSs also report increased concentrations of regulated organic contaminants and disinfection by-products.[44]

Our analyses stratified by sociodemographic subgroups relied on previously developed sociodemographic clusters and do not solely reflect racial or ethnic composition or socioeconomic status. Future analyses should comprehensively and specifically evaluate the associations of social vulnerability, socioeconomic status, and racial or ethnic composition

with CWS metal concentrations, which was beyond the scope of this analysis. Although we consistently found that 1% increases in the proportion of Hispanic/Latino residents were associated with higher contaminant metal exposure in overall and regional analyses, region-specific associations might exist for other racial or ethnic groups and CWS metals. We found significantly higher chromium concentrations for CWSs serving correctional facilities versus all CWSs, and one previous analysis found elevated CWS arsenic concentrations for incarcerated populations in the Southwestern USA.[7] Future analyses could specifically evaluate CWS metal exposures for incarcerated populations by region and by racial or ethnic composition.

Even at low concentrations, uranium, a naturally occurring radioactive metal, represents an important risk factor for the development of chronic diseases.[45] Despite the potential health effects of uranium exposure, little epidemiological research has been done on chronic, low-level water uranium exposures, especially in CWSs. Previous studies found associations between chronic uranium exposure and increased risk of hypertension, cardiovascular disease, kidney damage, and lung cancer.[45–47] Additional resources such as further compliance enforcement and increased technical and financial assistance to improve water treatment are needed to lower uranium concentrations in CWSs, especially in highly exposed communities.

Limitations regarding SYR3 data quality have previously been described in detail and should be considered.[6] Briefly, SYR record submission is voluntary and we are therefore missing records for a small number of CWSs (appendix 2 p 5). LOD reporting was not uniform, resulting in differing LODs across CWSs and missing LODs from several records. We were unable to aggregate CWS concentrations to more refined geographic resolutions (eg, census block, postal code) because CWS distribution boundaries are not available nationwide. Finally, reported CWS source water type in SDWIS (surface *vs* groundwater) might have changed over time and influenced our findings stratified by source water type.

Additional, specific limitations for uranium should also be considered. Although the present study shows that uranium concentrations and detections in CWSs are higher than those for other metals,[6,7] these findings might be biased by the EPA Standardized Monitoring Framework for uranium.[48] CWS concentration estimates for uranium were derived by combining compliance monitoring records from both SYR2 (2000–05) and SYR3 (2006–11). CWSs were required by the EPA Framework to conduct initial monitoring between 2000 and 2007 (covering the collection of grandfathered data and the initial compliance monitoring period for radionuclides), and the EPA First Radionuclides Rule Compliance Cycle covers the period 2008–16. CWSs with uranium concentrations below the LOD during the initial compliance monitoring period were only required by the Framework to have one sample collected during the First Radionuclides Rule Compliance Cycle (2008–16). Consequently, not all CWSs collected a compliance monitoring sample during the period that we examined (2000–11), resulting in a smaller number of CWSs included in our uranium analysis, and potentially biased concentration estimates (potentially overestimating nationwide uranium CWS concentrations and MCL exceedances) due to differential missingness for CWS uranium concentrations. Furthermore, national spatial coverage for uranium was poor compared with coverage for the other metals we examined.

Particular regions, such as the Eastern Midwest, Southeast, Mid-Atlantic, and New England, have noticeably poor spatial coverage for CWS uranium concentrations. Although CWS uranium estimates were largely similar when averaging to different periods (appendix 2 p 9), incorporating data from the fourth SYR once available (covering 2012–17) will potentially improve concentration estimates and is needed to confirm disproportionately exposed communities. Given the similar estimates across multiple periods, we assumed in this study that uranium concentrations did not decrease over time (2000–11), although further analyses with data from the fourth SYR are needed. Although we could not account for reported treatment in SYR2 records, the uranium MCL was not yet in effect and treatment to reduce uranium concentrations was unlikely.

The present study indicates that 2·1% of CWSs with data available report average uranium concentrations (2000–11) in exceedance of the EPA MCL, and that uranium is frequently detected during compliance monitoring. Arsenic, barium, chromium, selenium, and uranium concentrations are disproportionately elevated in CWSs serving Semi-Urban, Hispanic populations, raising environmental justice concerns for these communities and the possibility that inequalities in public drinking water metal exposures are influencing inequalities in several metal-associated disease outcomes, including diabetes, liver disease, and cardiovascular disease. Additional regulatory policies, compliance enforcement, and improved infrastructure are therefore necessary to reduce disparities in CWS metal concentrations and protect communities served by public water systems with elevated metal concentrations. Such interventions and policies should specifically protect the most highly exposed communities to advance environmental justice and protect public health.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Data sharing

An online dashboard containing an interactive map and searchable table of metal concentration estimates at the community water system (CWS) and county levels are available online. Datasets of the average metal concentrations at the CWS and county levels are available in appendix 3 and online.

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Research in context

Evidence before this study

We searched PubMed on Feb 1, 2022 for peer-reviewed articles containing “uranium” AND “public water” OR “public drinking water”, with no language or publication year restrictions. No nationwide estimates have been published on uranium concentrations in US regulated public drinking water systems (serving >90% of US residents) that can be used for epidemiological purposes. An estimated 4% of private domestic water wells in the USA have uranium concentrations exceeding US Environmental Protection Agency (EPA) maximum contaminant levels (MCL) of 30 μg/L, suggesting uranium might also be widespread in public water systems. Furthermore, previous studies characterised significant sociodemographic inequalities in concentrations of arsenic and nitrates in US public water, through use of routine compliance monitoring records compiled by EPA, suggesting substantial environmental injustices might also exist in exposure to uranium and other metals in public drinking water.

Added value of this study

We developed nationwide estimates of uranium and nine other regulated metals (antimony, arsenic, barium, beryllium, cadmium, chromium, mercury, selenium, and thallium), in community water systems (CWSs) across the USA using compliance monitoring records, which can be utilised in future epidemiological studies. We also identified significant sociodemographic inequalities in public water uranium concentrations. We found the highest estimated CWS uranium concentrations for Semi-Urban, Hispanic communities and communities located in the Southwest and Central Midwest regions. In a hierarchical cluster analysis, uranium, selenium, and arsenic clustered together, possibly reflecting groundwater redox conditions.

Implications of all the available evidence

Uranium is an underappreciated contaminant in US public drinking water systems, with 63·1% of available records reporting uranium detections and 2·1% of CWS averages (2000–11) exceeding the uranium MCL. Inequalities in CWS concentration estimates for Hispanic communities persisted after adjustment for potential confounders, suggesting inequalities possibly result from regulatory failure to protect marginalised communities and not from local geological context. Future epidemiological studies should explore the association between sociodemographic inequalities in public water metal concentrations and related adverse health outcomes, considering the environmental justice concerns highlighted in this study. Additional regulatory oversight and technical and financial assistance are needed for water systems serving highly exposed communities.

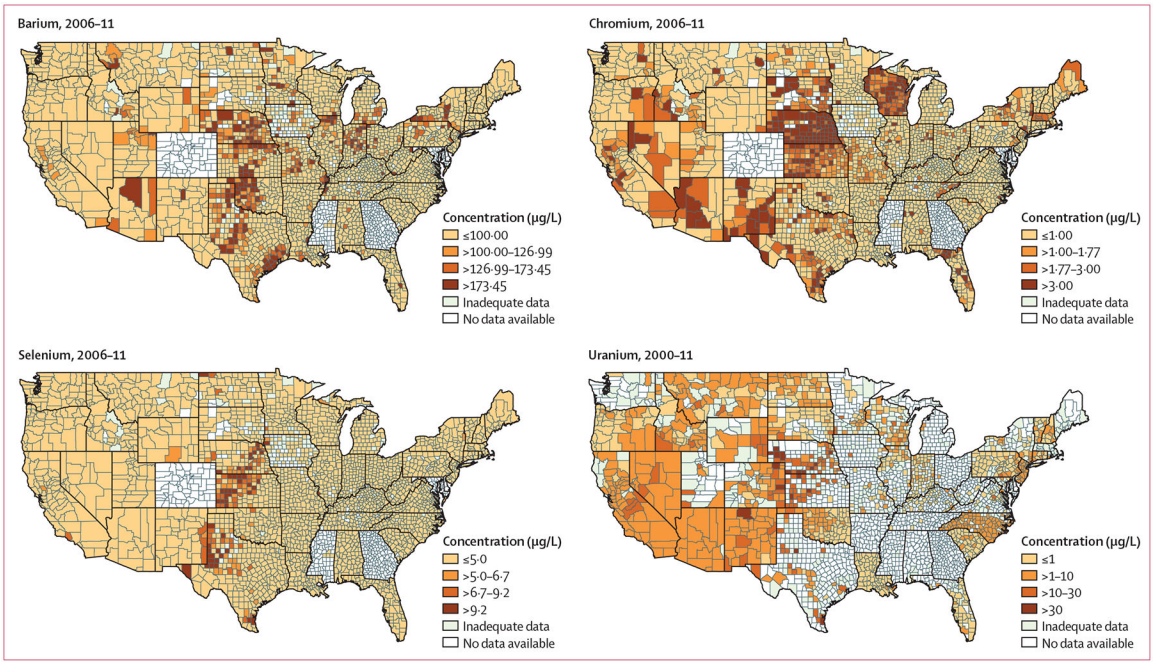

Figure 1: County-level weighted average of water contaminant concentrations in CWSs (n=37 915) from 2006–11 for barium (A), chromium (B), selenium (C), and uranium (D; 2000–11)

Average concentrations were weighted by the population served by each CWS to estimate the county-level weighted average CWS concentrations. Counties which were not represented by any CWSs in the SYR3 database are labelled as having no data available. Counties with inadequate data did not have CWS data representing at least 50% of the public water reliant population (appendix 2 pp 5–6). Estimates for uranium are derived from both the second (2000–05) and third (2006–11) SYR. For barium, chromium, and selenium, the lowest concentration category corresponds to less than or equal to the SYR3 minimum reporting level (100 µg/L for barium, 1 µg/L for chromium, 5 µg/L for selenium), and the other three categories reflect tertiles of the remaining distribution of county-level estimates. For uranium, the lowest concentration category corresponds to less than or equal to 1 µg/L, and the other three categories reflect cut-points that might be considered in future regulatory decisions. CWS=community water system. SYR=Six-Year Review.

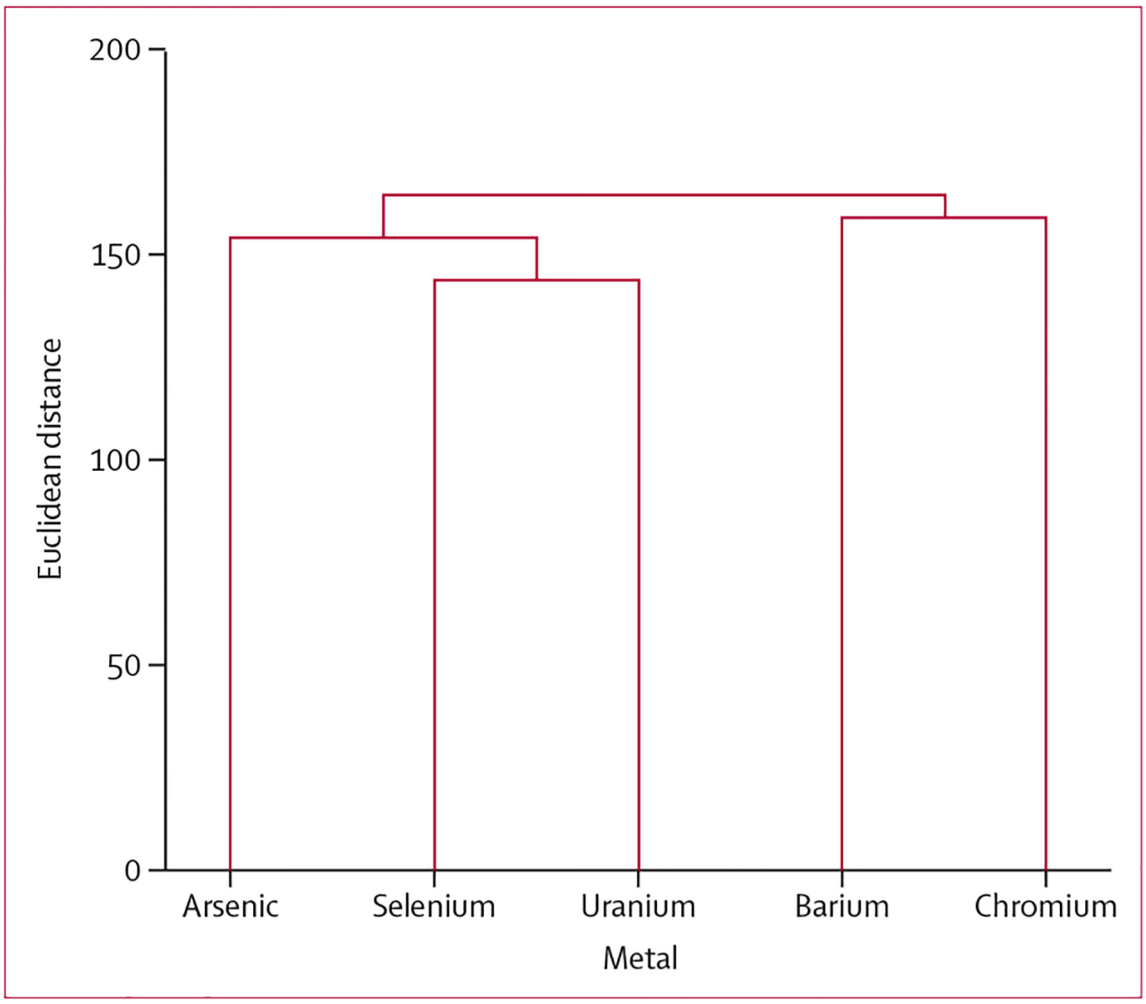

Figure 2: Dendrogram of hierarchical cluster analysis for 6-year averages (2006–11) of regulated metal concentrations in CWSs across the USA (n=12 756).

Analysis was restricted to CWSs with no missing concentration estimates for arsenic, selenium, uranium, barium, and chromium. Values for uranium are averaged from 2000–11. We used Ward's method for Euclidean distances and normalised each metal to unit variance and zero mean before analysis. CWS=community water system.

Table 1:

Distribution of metals in CWSs across the USA, 2006–11

	Antimony	Arsenic	Barium	Beryllium	Cadmium	Chromium (total)	Mercury (inorganic)	Selenium	Thallium	Uranium (2000–11)
Number of CWSs (n=37 915)	34 383	36 798	34 371	34 109	34 380	34 347	34 359	34 365	34 380	14 503
Number of records, 1000s	165	297	165	164	165	167	164	165	164	128
Number of detections (%) *	3635 (2·2%)	135 227 (45·5%)	100 521 (60·8%)	2165 (1·3%)	2623 (1·6%)	31 621 (18·9%)	2499 (1·5%)	21 422 (12·9%)	2605 (1·6%)	80 571 (63·1%)
Arithmetic mean concentration, µg/L	<0·4	1·77	66	<0·20	0·08	1·12	<0·20	1·11	<0·30	4·4
50th percentile, µg/L	<0·4	<0·5	25	<0·20	<0·05	<0·08	<0·20	<0·60	<0·30	1·0
75th percentile, µg/L	<0·4	1·66	79	<0·20	<0·05	0·62	<0·20	<0·60	<0·30	3·1
90th percentile, µg/L	<0·4	4·66	175	<0·20	0·06	3·00	<0·20	2·00	<0·30	10·9
95th percentile, µg/L	0·42	7·40	253	0·25	0·21	5·30	<0·20	3·81	<0·30	18·5
WHO GDWQ limit, µg/L	20	10	1300	NA	3	50	6	40	NA	30
Number CWSs (%) > GDWQ limit	0 (0%)	953 (2·6%)	49 (0·1%)	NA	20 (0·1%)	18 (0·1%)	0 (0%)	28 (0·1%)	NA	299 (2·1%)
US EPA MCL, µg/L	6	10	2000	4	5	100	2	50	2	30
Number CWSs (%) > MCL	10 (<0·1%)	953 (2·6%)	9 (<0·1%)	8 (<0·1%)	8 (<0·1%)	4 (<0·1%)	4 (<0·1%)	15 (<0·1%)	10 (<0·1%)	299 (2·1%)

All concentration estimates (excluding uranium) are based on EPA SYR3 records and cover 2006–11; uranium concentration estimates are based on EPA SYR2 and SYR3 records and cover 2000–11. Values lower than the EPA maximum limit of detection for each metal are displayed as < the value of the detection limit in µg/L. CWS=community water system. GDWQ=Guidelines for Drinking-water Quality. EPA=Environmental Protection Agency. MCL=maximum contaminant level. SYR=Six-Year Review.

* Percentages calculated with unrounded denominators.

Table 2:

75th and 95th percentiles of metal concentrations in CWSs nationwide and stratified by subgroup (n=37 915, 2006–11)

	Arsenic		Barium		Chromium		Selenium		Uranium (2000–11)	
	n	75th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L	n	75 th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L
All CWSs	36 798	1·66, 7·40	34 371	79, 253	34 347	0·62, 5·30	34 365	<0·60, 3·81	14 503	3·1, 18·5
Source water type										
Groundwater *	33 155	1·86, 7·77	30 863	87, 265	30 838	0·70, 5·59	30 858	<0·60, 4·10	12 996	3·4, 19·5
Surface water	3643	0·63, 2·39	3508	40, 107	3509	0·27, 2·36	3507	<0·60, 2·10	1507	1·5, 7·1
p value	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	0·43	NA	<0·0001
Size of population served †										
≤500	21 269	1·90, 8·08	19 458	76, 255	19 442	0·53, 5·56	19 454	<0·60, 3·89	8565	3·5, 20·7
501–3300	8839	1·58, 6·94	8372	94, 275	8370	0·76, 5·43	8375	<0·60, 4·38	3136	2·9, 17·9
3301–10 000	3428	1·43, 5·94	3323	83, 240	3318	0·62, 4·53	3318	0·68, 3·56	1293	2·3, 14·7
10 001–100 000	2877	1·16, 4·96	2836	58, 186	2835	0·71, 4·35	2836	0·75, 3·01	1259	2·9, 11·2
>100 000	385	1·10, 4·19	382	44, 120	382	0·84, 3·28	382	0·81, 2·73	250	3·4, 11·0
p value	NA	0·0020	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	<0·0001
Region ‡										
Alaska and Hawaii	486	1·17, 9·53	429	37, 141	429	1·10, 3·63	428	<0·60, 1·56	346	0·7, 1·6
Central Midwest	2655	2·64, 7·23	2609	149, 304	2609	3·10, 8·69	2609	2·71, 12·50	797	11·4, 32·2
Eastern Midwest	6085	2·15, 7·49	5714	111, 309	5712	<0·08, 5·88	5712	1·01, 3·54	1395	1·0, 3·7
Mid-Atlantic	4902	0·35, 3·32	3809	97, 338	3798	<0·08, 2·78	3805	<0·60, 1·16	2367	2·2, 9·9
New England	1728	2·10, 6·95	1702	20, 75	1702	1·02, 3·54	1700	<0·60, 2·52	736	4·0, 25·1
Pacific Northwest	4456	2·50, 8·50	3840	27, 150	3839	<0·08, 1·90	3842	<0·60, 1·01	1668	2·7, 16·0
Southeast	7866	0·35, 2·00	7765	21, 130	7764	<0·08, 2·54	7763	<0·60, 1·46	3980	2·1, 7·8
Southwest	8617	3·20, 10·73	8503	115, 270	8494	1·49, 7·68	8506	1·03, 6·50	3214	9·6, 28·3
p value	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	<0·0001
Sociodemographic county cluster §										
Semi-Urban, High SES	14 604	1·41, 6·30	13 330	75, 250	13 313	0·28, 4·64	13 323	<0·60, 3·08	6062	2·3, 15·8
Semi-Urban, Middle-to-Low SES	1409	0·35, 1·58	1405	16, 107	1404	<0·08, 1·30	1403	<0·60, 0·71	862	1·0, 2·8

	Arsenic		Barium		Chromium		Selenium		Uranium (2000–11)	
	n	75th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L	n	75 th, 95th percentiles, µg/L	n	75th, 95th percentiles, µg/L
Semi-Urban, Hispanic	4635	3·60, 11·71	4555	110, 279	4558	1·49, 8·00	4557	1·27, 8·49	2045	10·9, 31·7
Mostly Rural, Middle SES	9100	0·86, 6·00	8434	64, 240	8429	0·53, 4·10	8433	<0·60, 2·59	2803	2·0, 9·5
Rural, Middle-to-Low SES	533	0·73, 4·50	526	62, 198	523	0·59, 5·00	526	<0·60, 4·29	194	2·0, 14·7
Young, Urban, Middle-to-High SES	1038	2·67, 9·35	1006	110, 227	1010	0·65, 6·97	1009	<0·60, 3·79	538	6·3, 17·7
Rural, American Indian	486	3·17, 10·17	444	90, 252	444	0·98, 5·35	448	0·74, 4·85	382	2·7, 10·3
Rural, High SES	5188	2·55, 8·08	4853	100, 266	4851	1·38, 6·35	4851	0·82, 7·21	1716	3·7, 22·2
p value	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	<0·0001	NA	<0·0001
Correctional facility CWSs[¶]	203	2·48, 9·22	192	56, 227	191	1·33, 7·04	192	0·62, 3·12	74	3·2, 12·4
p value	NA	0·13	NA	0·35	NA	0·042	NA	0·28	NA	0·64

All concentration estimates (excluding uranium) are based on EPA SYR3 records and cover 2006–11; uranium concentration estimates are based on EPA SYR2 and SYR3 records and cover 2000–11. p values are from non-parametric Kruskal-Wallis tests (comparing the distribution of concentrations across subgroup categories). CWS=community water system. NA=not available. SES=socioeconomic status. EPA=Environmental Protection Agency. SYR=Six-Year Review.

[*] CWSs served by groundwater include those served by surface water under the influence of groundwater and groundwater under the influence of surface water.

[†] Standard US EPA categories; population served is the adjusted total population served (as defined by EPA), which accounts for systems that sell or purchase water and avoids overcounting.

[‡] States included in geological regions are: Alaska and Hawaii (AK, HI), Central Midwest (ND, SD, NE, KS, MO), Eastern Midwest (WI, IL, IN, MI, OH, MN, IA), Mid-Atlantic (PA, MD, DC, DE, NY, NJ, CT, RI), New England (MA, VT, NH, ME), Pacific Northwest (WA, OR, MT, WY, and ID), Southeast (OK, AR, LA, MS, AL, FL, GA, TN, KY, SC, NC, VA, WV), and Southwest (CA, NV, UT, CO, AZ, NM, TX).

[§] 143 CWSs served more than one county; of these, approximately half served counties categorised to different sociodemographic county-clusters (eg, the CWS NY7003493 serves New York, NY [Young, Urban, Middle-to-High SES] and Bronx, NY [Semi-Urban, Hispanic]); sociodemographic clusters were classified on the basis of Wallace et al;[23] the CWSs are represented for each county that they serve in the sociodemographic county-cluster analyses (n=36 674 CWSs analysed in sociodemographic county-cluster analyses).

[¶] CWSs exclusively serving correctional facilities were identified via a keyword search of CWS names for “prison”, “correction”, “corr”, “juvenile”, “detention”, “jail”, “penitentiary”, “women”, “TDCJ” (Texas Department of Criminal Justice), “ADOC” (Arizona Department of Corrections), “ADC” (Arkansas Department of Correction), and “sheriff”. Values lower than the EPA maximum limit of detection for each metal are displayed as < the value of the detection limit in µg/L.

Table 3:

75th, 90th, and 95th percentiles of uranium concentrations in CWSs located in California, Oklahoma, and Texas stratified by Semi-Urban, Hispanic counties versus all other counties

	CWSs serving Semi-Urban, Hispanic counties		All other CWSs	
	Number of CWSs	Uranium, 75th, 90th, and 95th percentiles, μg/L	Number of CWSs	Uranium, 75th, 90th, and 95th percentiles, μg/L
California	822	11·7, 23·5, 35·1	722	6·4, 13·0, 18·3
Oklahoma	16	11·4, 35·6, 49·0	571	1·9, 6·4, 11·3
Texas	167	20·6, 33·3, 44·6	105	10·5, 20·2, 24·5

CWS=community water system.

Table 4:

90th percentile difference in CWS uranium concentration per 1% higher proportion of county populations classified as Hispanic/Latino

	Number of CWSs	Beta
All CWSs	14 644	15·1 (p<0·0001)
CWSs in the Southwest region	3268	25·9 (p<0·0001)
CWSs serving Semi-Urban, Hispanic counties	2058	11·2 (p=0·22)

Model results for the difference in CWS 90th percentile uranium concentrations per 1% higher proportion of population classified as Hispanic/Latino were derived from quantile regression models with the quantreg package (version 5.85) in R. Models were adjusted for state (categorical), the size of the population served by the CWS (continuous), and the source water type (surface water *vs* groundwater), and SEs were bootstrapped. The proportion of the county population classified as Hispanic/Latino was estimated for the 2010–19 period with use of US Census Bureau statistics.[24,25] CWS=community water system.