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Supplementary appendix 2

This appendix formed part of the original submission and has been peer reviewed. We post it as supplied by the authors.

Supplement to: Ravalli F, Yu Y, Bostick BC, et al. Sociodemographic inequalities in uranium and other metals in community water systems across the USA, 2006–11: a cross-sectional study. *Lancet Planet Health* 2022; **6:** e320–30.

Appendix

Ravalli et al. 2022, Sociodemographic inequalities in uranium and other metals in community water systems across the US, 2006-2011: a cross-sectional study

Appendix Methods: Supplemental details. Information not also included in the main manuscript text is underlined. References cited here are numbered in accordance with the main manuscript text, and full references are available at the end of this section.

EPA's database supporting the Second Six Year Review (SYR2) and Third Six Year Review (SYR3)

The US EPA compiles compliance monitoring data from public water systems for regulated drinking water contaminants every six years as required by the SDWA.[16,17] Data are voluntarily sent in from states, territories, and tribal authorities and EPA works directly with agencies to collect records for a given monitoring period. We used monitoring data from the Third Six Year Review (SYR3) period (2006-2011), which includes approximately 13 million total analytical records from 139,000 public water systems serving 290 million people annually. Data from 46 states, Washington D.C., and American Indian tribes (including those in EPA Regions 1, 4, 5, 8, and 9, and Navajo Nation) were included, representing 95% of all public water systems and 92% of the total population served by public water systems nationally.[16,17] In the SYR3 period, Colorado, Delaware, Georgia, Mississippi, and EPA Regions 2, 6, 7, and 10 (tribal systems) did not submit data.[16] EPA conducted extensive quality assurance and quality control assessments prior to publishing the final SYR3 data.[16-18]

To determine unbiased CWS concentration estimates for uranium, we also utilized uranium compliance monitoring records from the Second Six Year Review (SYR2) database which includes records from 2000-2005. Unlike MCLs for other metals regulated by EPA, the MCL for uranium (30 µg/L) was established in 2000 under the Radionuclides Final Rule. We included uranium in the current study of metals because uranium in drinking water is mostly natural uranium rather than enriched uranium, and natural uranium is related to adverse health outcomes (e.g. nephrotoxicity).[19] Public water systems were required to collect at minimum one

grandfathered/initial uranium monitoring sample between 2000 and 2007, which determined the frequency of compliance monitoring sample collection during the subsequent compliance monitoring cycle (2008-2016). For the other metals, all public water systems were required to collect at minimum one compliance monitoring sample every three-years in accordance with the Standard Monitoring Framework for inorganic contaminants, and thus SYR2 records were not needed to generate unbiased concentration estimates.[20] For all metals, records from water systems categorized as transient or non-transient non-community water systems were excluded because our focus was on CWSs which serve the large majority of the US population year round (>90%) and only CWSs are subject to the Radionuclide Rule for uranium.[16]

Metal concentrations below the limit of detection (LOD) were replaced by the record-specific LOD divided by the square root of two (this method is used by the Centers for Disease Control and Prevention and other federal agencies when reporting geometric or arithmetic means of environmental biomarkers and concentration estimates).[21] Although EPA has established method-specific maximum LODs for these metals, many water systems reported monitoring records with lower and higher LODs than the EPA established maximum LOD, or did not report record-specific LODs at all. This reflects the challenges related to the voluntary nature of the SYR system. For these records, we imputed the value of the EPA's method detection limit divided by the square root of two for any non-detect record that did not report a record-specific LOD in mg/L or µg/L, or which reported a LOD greater than 5 µg/L, as these were considered unreliable. The percentage of records with values above the detection limit 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. Since only 5 metals (arsenic, barium, chromium, selenium, and uranium) had over 10% of

records above the LOD, we restricted our main analyses to these metals. We excluded records reporting concentrations greater than 100 times the MCL, as these are likely errors (N= 2 records for selenium, 2 for thallium, and 6 for uranium). Some records with concentration values greater than 10 µg/L were reported as "non-detect" records (N=28 for uranium records from the SYR2, and N=161 for uranium records from the SYR3). Excluding these samples resulted in similar findings (results not shown).

Statistical analysis: CWS-level metal concentration estimates

For each metal, many CWSs reported multiple monitoring records per year and compliance monitoring period. We averaged available monitoring records for each metal at the CWS-level to 2006-2011 (the overall time period for records in the SYR3). As a sensitivity analysis, we also averaged records to 2008-2010 (the time period corresponding to EPA's Standard Monitoring Framework for inorganic contaminants[20]), with similar findings (**appendix page 8**). For uranium, we averaged CWS-level uranium concentrations to the 2000-2011 time period, which covers grandfathered/initial compliance samples (2000-2007) and samples collected during the first compliance monitoring cycle (2008-2016). Because the 2000-2011 time period covers the grandfathered/initial and incompletely covers the first compliance monitoring cycle, we conducted a sensitivity analysis assessing the distribution of CWS-level uranium concentrations when averaged to 2000-2007 (the grandfathered/initial compliance time period), 2008-2011 (incomplete first compliance monitoring cycle) and 2006-2011 (the overall time period for records in the SYR3), also with similar findings (**appendix page 9**).

We accounted for reported treatment within each calendar year before averaging CWS concentrations to the overall averaging time periods. Although detailed treatment methods are

not available for most individual CWS, general treatment methods include coagulation, sedimentation, filtration, and disinfection.[22] Few CWSs reported records for both "raw" and "finished" (i.e. treated) water samples within the same year. When the average concentration of metals in finished water samples was lower than in raw samples, we calculated the yearly average only with the finished water samples. Because this information was not available in the SYR2 and the MCL for uranium did not become enforceable until 2008, average yearly uranium estimates only accounted for treatment in records from 2006-2011.

To assign counties-served for each CWS, we merged the monitoring data for each metal 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 (surface versus groundwater)[23] as previously described in detail.[7] A small number of CWSs that were missing from the SDWIS database could not be merged directly with SDWIS and were excluded from analysis (N= 31/34,414 for antimony, 32/34,403 for barium, 39/34,148 for beryllium, 30/34,410 for cadmium, 24/34,371 for chromium, 28/34,387 for mercury, 28/34,393 for selenium, 32/34,412 for thallium, and 85/9,835 for uranium).

County-level maps of CWS-level metal concentration estimates

To visually identify spatial patterns in metal concentration estimates across the US, we also estimated county-level CWS metal concentrations, as previously described in detail.[7] Briefly, we estimated county-level average metal concentrations weighted by the number of people served by each CWS within a county (population served) for both time periods (2000-2011 for uranium, and 2006-2011 for all other metals). CWSs not reporting data for a particular period did not contribute to that county's total population served or average. To avoid reporting a

county-level average derived from CWSs which only served small populations relative to the entire county population, the average water metal concentration for a given county was only estimated if the CWSs serving that county reported serving at least fifty percent or more of the public water-reliant population in the entire county (few counties were treated as missing for this reason, and the percent of the population reliant on public water has increased over the past several decades). We estimated the public water-reliant population for each county using the latest nation-wide Census statistic on county-level household tap water source from the 1990 Census.[26,29] Because only county-served was reliably reported in SDWIS for each CWS, we did not aggregate to smaller geographic scales (e.g. census-tract). We mapped the full county-level estimates of water metal averages across the conterminous US using the *maps* package version 3.3.0 in R.[30]

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Supplemental Table 1. Distribution of inorganic contaminants (except uranium) in community water systems (CWSs) across the US in µg/L, averaged to either 2006-2011 (entire time period covering the Third Six Year Review) or 2008-2010 (corresponding to the US EPA's Standard Monitoring Framework compliance monitoring period). Estimates for uranium when averaged to different time periods are presented in Supplemental Table 2.

	Antimony	Arsenic	Barium	Beryllium	Cadmium	Chromium (total)	Cyanide	Mercury (inorganic)	Selenium	Thallium
2006-2011										
N CWSs	34,383	36,798	34,371	34,109	34,380	34,347	24,664	34,359	34,365	34,380
Arithmetic mean	0.32	1.77	66.32	0.16	0.08	1.12	4.11	0.14	1.11	0.22
50%	0.28	0.35	24.52	0.14	0.04	0.06	3.54	0.14	0.42	0.21
2008-2010										
N CWSs	28,437	32,127	28,288	28,356	28,307	28,269	19,929	28,269	28,281	28,430
Arithmetic mean	0.32	1.78	62.81	0.16	0.08	1.06	3.98	0.14	1.12	0.22
50%	0.28	0.35	21.14	0.14	0.04	0.06	3.54	0.14	0.42	0.21

Appendix Table 2. Distribution of uranium in community water systems in the US when averaged to three different time periods. The first time period (2000-2007) corresponds to the period for collecting grandfathered samples or initial monitoring samples for the Final Radionuclides Rule.

	2000-2007	**2008-2011**	**2000-2011**
N CWSs	9,750	8,805	14,503
Arithmetic mean	5.41	4.61	4.37
50%	1.48	0.71	1.00
90%	13.00	12.00	10.85
95%	22.35	20.00	18.54
N (%) above WHO GL and EPA MCL (30 µg/L)	304 (3.1%)	166 (1.9%)	299 (2.1%)

WHO = World Health organization. GDWQ = Guideline for drinking water quality. US EPA = US Environmental Protection Agency. MCL = maximum contaminant level.

Appendix Table 3. Arithmetic mean (95% CI) of metal concentrations (µg/L) in community water systems (CWSs) nationwide and stratified by subgroup (2006-2011).

	Barium		Chromium		Selenium		Uranium 2000-2011	
	N	**Mean (95% CI)**	**N**	**Mean (95% CI)**	**N**	**Mean (95% CI)**	**N**	**Mean (95% CI)**
All CWSs	34,371	66.3 (63.4, 69.2)	34,347	1.12 (1.00, 1.23)	34,365	1.11 (1.07, 1.14)	14,503	4.37 (4.05, 4.70)
Source water type								
Groundwater[a]	30,863	70.1 (66.9, 73.4)	30,838	1.19 (1.06, 1.31)	30,858	1.15 (1.11, 1.19)	12,996	4.67 (4.31, 5.04)
Surface water	3,508	32.7 (30.2, 35.2)	3,509	0.51 (0.44, 0.58)	3,507	0.73 (0.69, 0.76)	1,507	1.79 (1.59, 1.98)
Size of population served[b]								
≤500	19,458	63.8 (62.1, 65.6)	19,442	1.17 (0.99, 1.36)	19,454	1.13 (1.08, 1.19)	8,565	5.04 (4.51, 5.57)
500-3,300	8,372	79.7 (68.7, 90.8)	8,370	1.04 (0.98, 1.10)	8,375	1.16 (1.11, 1.22)	3,136	3.89 (3.50, 4.28)
3,301-10,000	3,323	65.7 (62.0, 69.3)	3,318	1.14 (0.69, 1.59)	3,318	0.99 (0.92 ,1.06)	1,293	3.01 (2.64, 3.37)
10,001-100,000	2,836	48.6 (45.7, 51.5)	2,835	0.94 (0.85, 1.04)	2,836	0.92 (0.86, 0.98)	1,259	2.77 (2.50, 3.05)
>100,000	382	36.1 (27.7, 44.4)	382	0.87 (0.62, 1.11)	382	0.98 (0.74, 1.23)	250	2.71 (2.26, 3.17)
Region								
Alaska/Hawaii	429	33.7 (27.4, 40.1)	429	0.93 (0.79, 1.08)	428	0.68 (0.58, 0.78)	346	0.89 (0.68, 1.10)
Central Midwest	2,609	108.2 (104.3, 112.1)	2,609	2.40 (2.28, 2.53)	2,609	2.77 (2.55, 2.99)	797	8.04 (7.01, 9.07)
Eastern Midwest	5,714	92.5 (88.8, 96.2)	5,712	1.01 (0.94, 1.08)	5,712	1.00 (0.96, 1.04)	1,395	1.24 (1.09, 1.40)
Mid-Atlantic	3,809	104.2 (80.0, 128.5)	3,798	1.53 (0.54, 2.53)	3,805	0.57 (0.54, 0.6)	2,367	2.92 (2.47, 3.37)
New England	1,702	17.8 (16.2, 19.5)	1,702	0.73 (0.68, 0.79)	1,700	0.69 (0.65, 0.73)	736	6.59 (5.18, 8.01)
Pacific Northwest	3,840	30.0 (27.6, 32.3)	3,839	0.37 (0.31, 0.43)	3,842	0.61 (0.57, 0.66)	1,668	3.33 (2.94, 3.72)
Southeast	7,765	28.1 (26.1, 30.0)	7,764	0.50 (0.46, 0.55)	7,763	0.67 (0.63, 0.71)	3,980	2.09 (1.87, 2.31)
Southwest	8,503	81.6 (79.2, 84.0)	8,494	1.59 (1.50, 1.67)	8,506	1.64 (1.53, 1.75)	3,214	9.13 (7.81, 10.44)
Sociodemographic county cluster[c]								
Semi-Urban, High SES	13,330	68.3 (61.3, 75.4)	13,313	1.12 (0.85, 1.40)	13,323	0.89 (0.85, 0.92)	6,062	3.50 (3.21, 3.78)
Semi-Urban, Mid/Low SES	1,405	26.3 (20.3, 32.3)	1,404	0.36 (0.26, 0.47)	1,403	0.53 (0.49, 0.56)	862	1.02 (0.88, 1.17)
Semi-Urban, Hispanic	4,555	80.8 (77.3, 84.4)	4,558	1.60 (1.49, 1.71)	4,557	2.03 (1.84, 2.22)	2,045	10.04 (8.16, 11.92)
Mostly Rural, Mid-SES	8,434	58.6 (56.1, 61.0)	8,429	0.90 (0.75, 1.05)	8,433	0.78 (0.75, 0.81)	2,803	2.19 (1.99, 2.40)

Rural, Mid/Low SES	526	51.1 (44.2, 58.1)	523	0.89 (0.72, 1.06)	526	1.06 (0.88, 1.24)	194	3.16 (2.09, 4.24)
Young, Urban, Mid/High SES	1,006	64.9 (59.3, 70.6)	1,010	1.23 (1.04, 1.42)	1,009	0.96 (0.83, 1.10)	538	6.87 (4.12, 9.63)
Rural, American Indian	444	66.0 (56.2, 75.8)	444	1.04 (0.80, 1.29)	448	1.25 (0.97, 1.52)	382	3.80 (1.42, 6.19)
Rural, High SES	4,853	73.7 (70.6, 76.9)	4,851	1.24 (1.16, 1.33)	4,851	1.60 (1.48, 1.71)	1,716	5.22 (4.61, 5.83)
Correctional facility CWSs	192	50.8 (39.6, 62.0)	191	1.34 (0.94, 1.74)	192	1.05 (0.73, 1.38)	74	2.62 (1.78, 3.45)

[a]CWSs served by groundwater include those served by surface water under the influence of groundwater and groundwater under the influence of surface water. [b]Categories of population served are standard U.S. EPA categories. Population served is adjusted total population served, which accounts for systems that sell or purchase water and avoids overcounting. A total of 143 CWSs served more than one county; of these, approximately half served counties categorized to different sociodemographic county-clusters (e.g., NY7003493 serves New York, New York (Young, Urban, Mid/High SES) and Bronx, New York (Semi-Urban, Hispanic). Sociodemographic clusters were classified based on Wallace et al. (2019) [31]. These CWSs are represented for each county that they serve in the sociodemographic county-cluster analyses (N = 36,674). States included in geologic regions are: Alaska/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).

Appendix Table 4. Arithmetic mean (95% CI) of uranium concentrations (µg/L) in community water systems (CWSs) located in California, Oklahoma, and Texas, stratified by counties classified as *Semi-Urban, Hispanic* versus all other counties, and average change in CWS uranium estimates per 1% higher proportion of the county population classified as Hispanic/Latino.

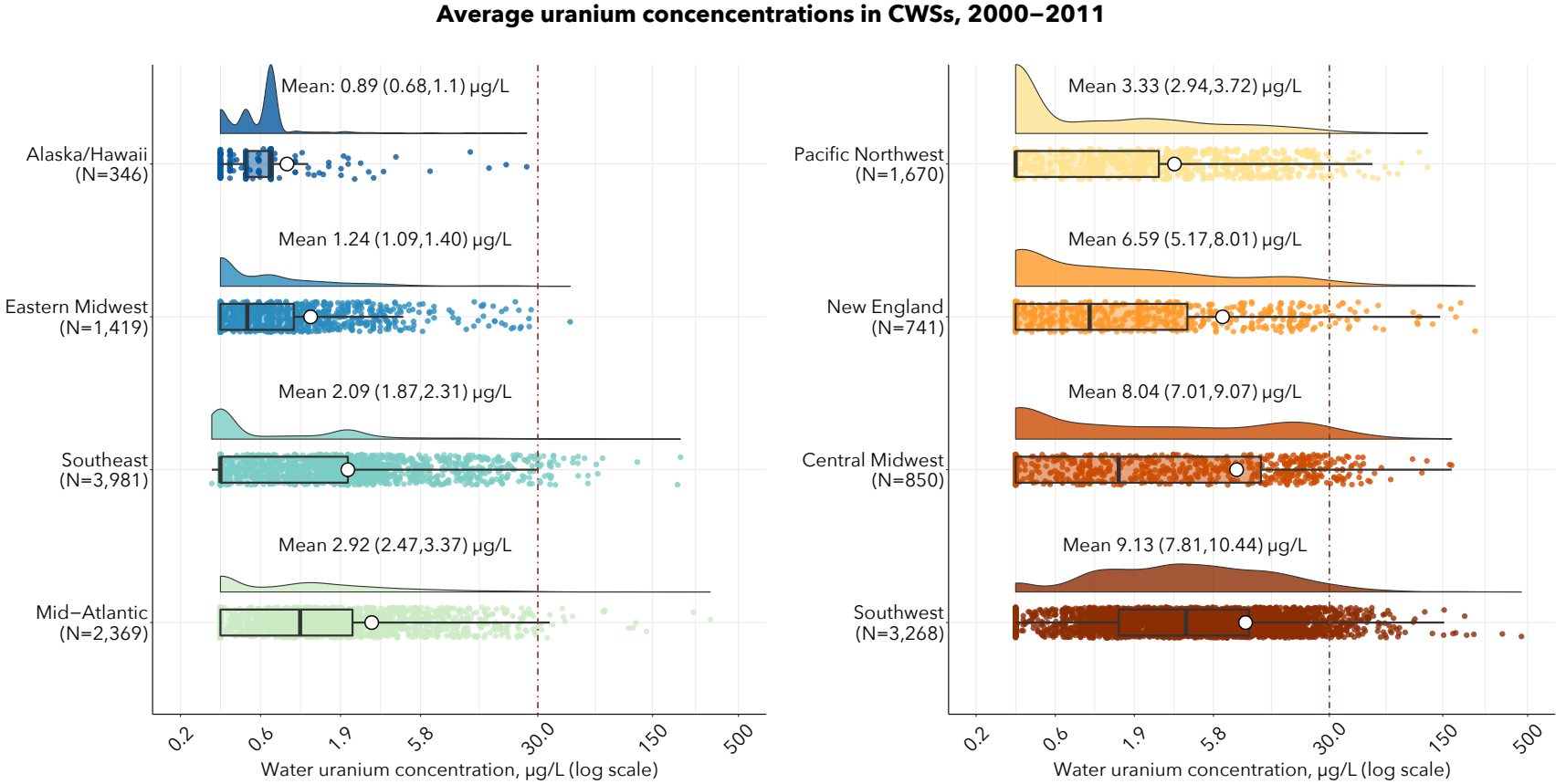
Arithmetic mean (95% CI) of CWS uranium concentrations

	CWSs serving *Semi-Urban, Hispanic*		**All other CWSs**	
	N CWSs	**Mean (95% CI) uranium**	**N CWSs**	**Mean (95% CI) uranium**
California	822	13.05 (8.43, 17.67)	722	6.45 (4.34 ,8.55)
Oklahoma	16	12.89 (6.3, 19.49)	571	2.45 (2.01, 2.89)
Texas	105	17.17 (14.4, 19.95)	105	8.04 (6.15, 9.93)

Arithmetic mean difference (p-value) in CWS uranium concentrations per 1% higher proportion of population classified as Hispanic/Latino

All CWSs		**CWS in the Southwest**		**CWSs serving *Semi-Urban, Hispanic***	
N CWSs	**Beta**	**N CWSs**	**Beta**	**N CWSs**	**Beta**
14,644	0.10 (p<0.001)	3,268	0.11 (p=0.01)	2,058	0.059 (p=0.53)

Appendix Figure 1. Distribution of average uranium concentrations (µg/L) in community water systems (CWSs) stratified by region for the period of 2000–2011. Filled polygons represent density plots. Box plot upper, middle, and lower hinges correspond to the 25th, 50th, and 75th percentiles, respectively. The 11-year average uranium concentration for each CWS is represented by a dot. Mean region-specific concentrations are indicated by the outlined white circle; mean (95% confidence intervals) are also listed for each region in text. The 30 µg/L maximum contaminant level is indicated by the red dashed line. The x-axis is truncated at 500 µg/L. The R code for this figure was adapted from: Allen M, Poggiali D, Whitaker K, Marshall TR, Kievit R. 2019. Raincloud plots: a multi-platform tool for robust data visualization. *Wellcome Open Res* 4:63, PMID: 31069261, https://doi.org/10.12688/wellcomeopenres.15191.1.


Average uranium concencentrations in CWSs, 2000–2011
Mean: 0.89 (0.68,1.1) µg/L
Alaska/Hawaii
(N=346)
Mean 1.24 (1.09,1.40) µg/L
Eastern Midwest
(N=1,419)
Mean 2.09 (1.87,2.31) µg/L
Southeast
(N=3,981)
Mean 2.92 (2.47,3.37) µg/L
Mid−Atlantic
(N=2,369)
0.2
0.6
1.9
5.8
30.0
150
500
Water uranium concentration, µg/L (log scale)
Mean 3.33 (2.94,3.72) µg/L
Pacific Northwest
(N=1,670)
Mean 6.59 (5.17,8.01) µg/L
New England
(N=741)
Mean 8.04 (7.01,9.07) µg/L
Central Midwest
(N=850)
Mean 9.13 (7.81,10.44) µg/L
Southwest
(N=3,268)
0.2
0.6
1.9
5.8
30.0
150
500
Water uranium concentration, µg/L (log scale)

Appendix Figure 2. Correlogram of Spearman's correlation coefficients between individual metal pairs for US community water systems (CWSs), 2006-2011. Uranium exposure estimates are from 2000-2011.