Supporting Information for

Can arsenic occurrence rates in bedrock aquifers be predicted?

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This supporting information includes 12 pages, 2 tables and 3 figures.

First, correlations between arsenic and hydrogeochemical parameters in groundwater samples are tabulated. Second, groundwater arsenic, pH, dissolved oxygen, nitrate and sulfate distributions in 2006 sampling from the greater Augusta area, Maine are plotted. Third, groundwater arsenic distribution in 2007 sampling is plotted. Fourth, the temporal variation of arsenic in well water between 2006 and 2007 sampling is presented. Fifth, results of logistic regression models are listed. Finally, a summary of documented bedrock aquifers with elevated arsenic of natural source is tabulated.

Table S1. Spearman's ρ values between arsenic and hydrogeochemical parameters in groundwater samples from Greater Augusta aquifers

Parameter	2006						2007			
	Greater Augusta	meta-sedimentary rocks			granite	volcanics	Chelsea	Sidney	Litchfield	Manchester
bedrock unit	all	Ss	Sw	SOv	D	OZc	D	Sw	Sw,SOv,D	Sw, D
n	790	161	259	248	86	30	107	62	49	113
well depth	0.21	0.15	0.28							
elevation							-0.26	-0.59		
temperature	0.09					0.57				
pH	0.54	0.50	0.52	0.49	0.65	0.44	0.48	0.50	0.64	0.43
dissolved oxygen	-0.35	-0.44	-0.29	-0.31	-0.41		-0.29	-0.43	-0.41	-0.36
Na					0.29	0.38				0.24
Mg	-0.11		-0.14						-0.35	
Si	-0.14		-0.22							
K	-0.12			-0.14					-0.56	
Ca	-0.08		-0.21						-0.64	-0.23
alkalinity					0.70					
chloride	-0.23		-0.26	-0.26	-0.21		-0.42		-0.48	-0.23
sulfate					0.24		-0.24			
nitrate	-0.31	-0.20	-0.36	-0.33	-0.24		-0.41	-0.45	-0.66	-0.48
cations minus chloride	0.10			0.14	0.38	0.44	0.22	0.34		0.22
fluoride	0.39	0.37	0.39	0.36	0.51		0.51	0.33	0.64	0.60
P	0.22	0.39	0.13	0.17			0.26		0.55	
V	0.16	0.20		0.12						
Cr		-0.17			-0.23		-0.27			
Mn	0.08		0.18	0.15			0.24			0.30
Fe							0.27			
Co	-0.20	-0.24	-0.23	-0.18	-0.40				-0.39	
Ni	-0.10		-0.20		-0.25			-0.42		
Cu	-0.24	-0.42	-0.19	-0.14	-0.37		-0.27			
Zn		-0.17			-0.35					
Rb								-0.33		
Sr			-0.21			0.38			-0.45	-0.23
Mo	0.38	0.29	0.34	0.45	0.48		0.46	0.67	0.61	0.61
Cd								0.39		0.27
In						0.37				
Sn					-0.24					
Sb	0.14			0.24						0.23
Cs	0.20	0.18	0.17	0.16	0.39					0.26
Ba	-0.28	-0.27	-0.28	-0.25	-0.25		-0.23		-0.52	-0.30
Re	-0.07							0.36		
Tl	0.15			0.21						
Pb	-0.22	-0.41	-0.16	-0.14	-0.41					
U	0.23	0.39	0.32	0.19	0.55		0.28	0.59		
Rn								-0.41		
cation-chloride vs. pH	0.17			0.32	0.57	0.61	0.49	0.30	0.33	

* Only values statistically significant ($p < 0.05$) are shown.

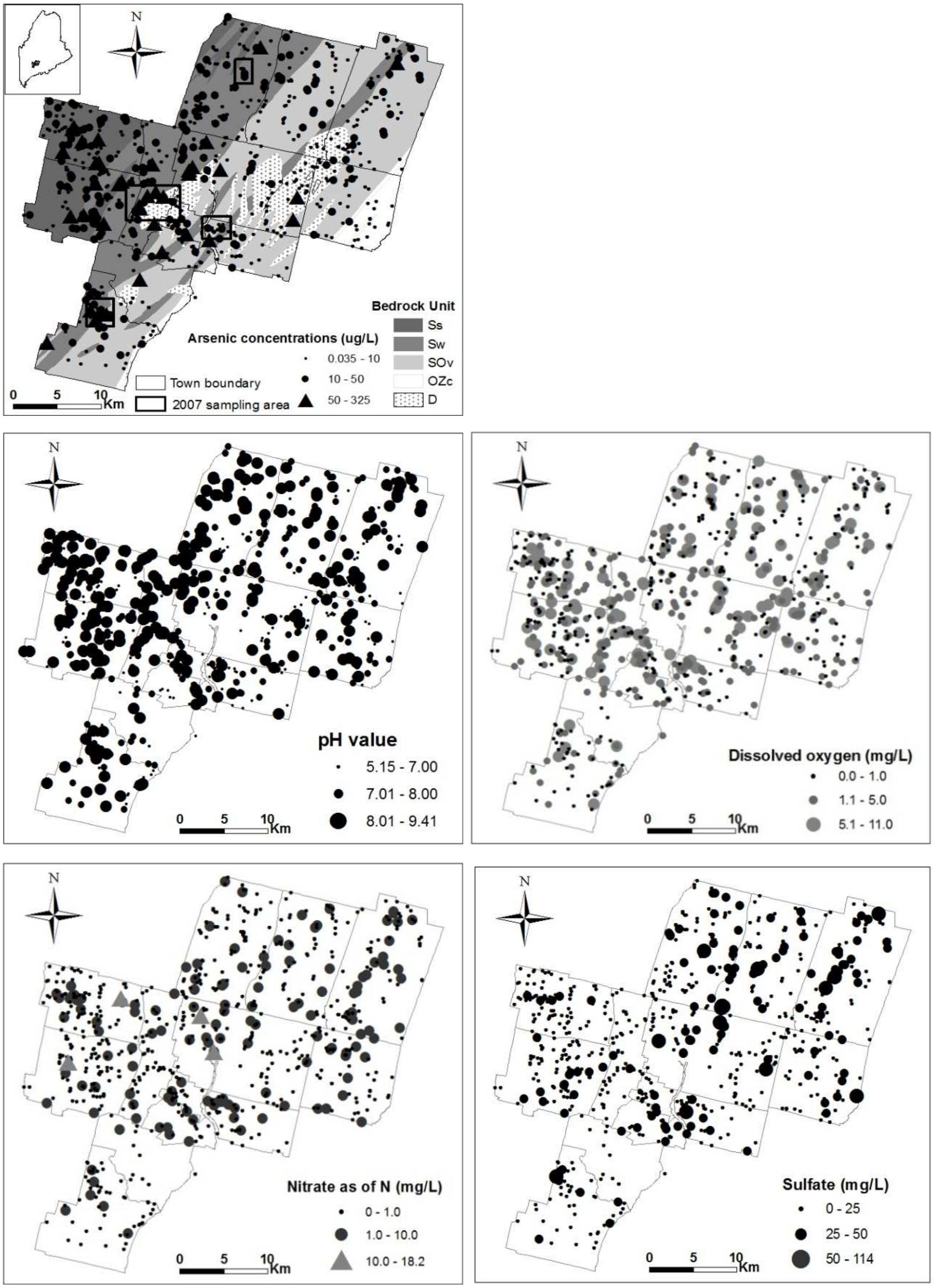
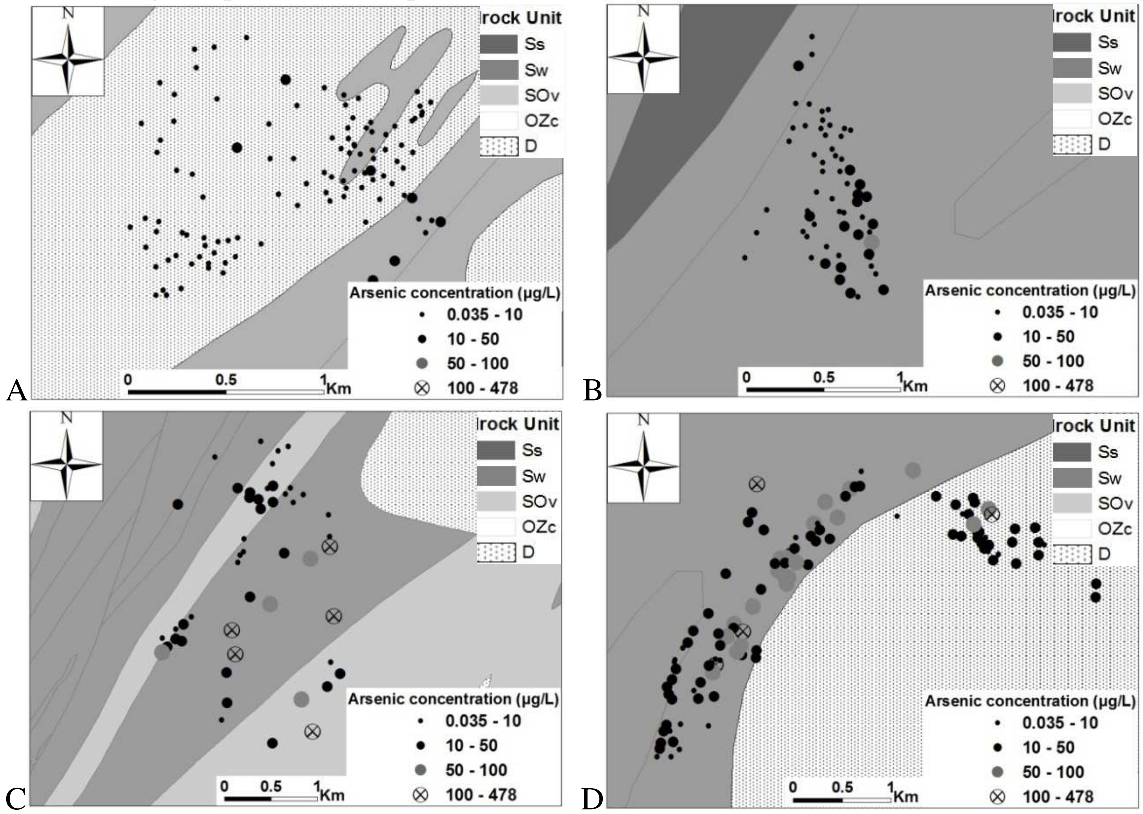


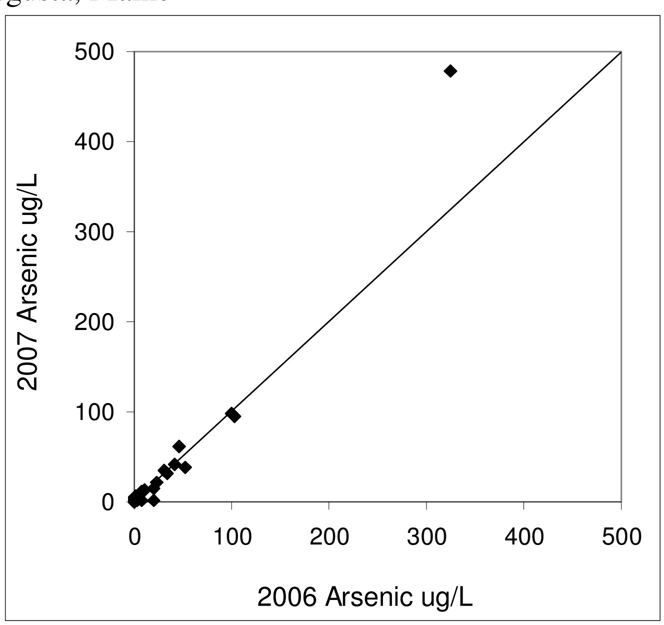
Figure S1. Groundwater arsenic distribution plotted on simplified bedrock geology map, and distributions of pH, dissolved oxygen, nitrate and sulfate in groundwater in 2006 sampling from Greater Augusta, Maine

Figure S2. Groundwater arsenic concentrations in 2007 sampling of four clusters in Greater Augusta plotted on simplified bedrock geology maps



A – Chelsea; B – Sidney; C – Litchfield; D – Manchester

Figure S3. Arsenic concentrations in groundwater sampled from same wells in 2006 and 2007 in Greater Augusta, Maine



* The solid line represents a ratio of 1:1.

Estimation of charge balance of major cations and anions in groundwater samples

In the 2006 sampling, 84 out of the 105 samples (or 80%) showed less than 20% relative difference between the molar sum of cations and the molar sum of anions. The formula to estimate the relative difference is as follows.

$$relative\ diference = \frac{sum\_of\_cations - sum\_of\_anions}{sum\_of\_cations + sum\_of\_anions} \times 100\%$$

This is probably because the alkalinity was measured one and a half year after the samples were stored in the lab and part of bicarbonate was lost. Actually in those 21 samples with greater than 20% relative difference, 20 of them had the molar sum of cations greater than the molar sum of anions.

In the 2007 sampling, which the alkalinity was measured 4-5 months after the samples were collected in the field, only 8 out of 331 samples (or 2%) showed greater than 20% relative difference between the molar sum of cations and anions.

Model results of stepwise logistic regression

The form of logistic regression model is $\frac{P[y=1|x]}{1-P[y=1|x]} = e^{(\beta_0+\beta_1 x_1+\beta_2 x_2+...+\beta_n x_n)}$. In this research, P[y=1|x] represents the response variable, probability of groundwater [As] >10 μg L^{-1}; x_1, x_2, … x_n represent explanatory variables or coefficients; β0 represents intercept, β1, β2, … βn represent estimate; standard error is the prediction error of each estimate; z value is estimate divided by standard error; Pr(>|z|) is the two-tailed p value, coefficients having p-values less than alpha (usually 0.05) are statistically significant.

The multivariate logistic regression model (including 12 parameters)

Coefficients	Estimate (β)	Std. Error	z value	Pr (>|z|)
(Intercept)	-8.999e+00	1.656e+00	-5.435	5.5e-08
Well Depth	-2.390e-06	9.525e-04	-0.003	0.99800
pH	9.698e-01	1.900e-01	5.106	3.3e-07
DO	-1.364e-01	6.448e-02	-2.115	0.03444
bedrock-OZ	1.545e-01	8.049e-01	0.192	0.84775
bedrock-S	1.464e+00	5.087e-01	2.879	0.00399
bedrock-SO	1.195e+00	5.223e-01	2.287	0.02217
Mg	5.342e-02	5.327e-02	1.003	0.31597
Si	1.838e-02	2.826e-02	0.650	0.51552
Ca	-5.374e-03	6.344e-03	-0.847	0.39699
F	-2.131e-01	2.816e-01	-0.757	0.44922
Cl	-1.270e-03	3.598e-03	-0.353	0.72397
NO3-N	-3.883e-01	1.820e-01	-2.133	0.03291
SO4	-2.646e-02	1.298e-02	-2.038	0.04154
Soil [As]	2.089e-02	1.355e-02	1.542	0.12316

Hosmer-Lemeshow goodness of fit test P value = 0.621
Deviance Residuals = -0.373

The final logistic regression model (including 5 parameters)

Coefficients	Estimate	Std. Error	z value	Pr (>|z|)
(Intercept)	-9.520490	1.324835	-7.186	6.66e-13
PH	1.075018	0.145576	7.385	1.53e-13
DO2_MGL	-0.117160	0.050389	-2.325	0.02007
bedrock-D	0.569743	0.713077	0.799	0.42430
bedrock-SOv	1.298917	0.650680	1.996	0.04591
bedrock-Ss	1.651966	0.656266	2.517	0.01183
bedrock-Sw	1.577493	0.647097	2.438	0.01478
NO3_NMGL	-0.239448	0.129868	-1.844	0.06521
SO4MGL	-0.028994	0.009454	-3.067	0.00216

Hosmer-Lemeshow goodness of fit test P value = 0.382
Deviance Residuals = -0.402
c statistic = 0.808

The oversimplified logistic regression model (including 4 parameters of water geochemistry only)

Coefficients	Estimate	Std. Error	z value	Pr (>|z|)
(Intercept)	-8.796945	1.174776	-7.488	6.98e-14
PH	1.160742	0.142049	8.171	3.05e-16
DO	-0.112959	0.049291	-2.292	0.021924
NO3_N	-0.203863	0.126519	-1.611	0.107109
SO4	-0.030703	0.008968	-3.423	0.000618

Deviance Residuals = -0.413
c statistic = 0.797

The oversimplified logistic regression model (including bedrock geology only)

Coefficients	Estimate	Std. Error	z value	Pr (>|z|)
(Intercept)	-2.2687	0.6064	-3.741	0.000183
bedrock-D	0.5296	0.6769	0.782	0.433986
bedrock-SOv	1.0821	0.6247	1.732	0.083222
bedrock-Ss	1.9773	0.6272	3.153	0.001618
bedrock-Sw	1.8442	0.6194	2.978	0.002906

Deviance Residuals = -0.730
c statistic = 0.636

Bedrock formations, Ss, Sw, SOv, D and OZc, were considered as one parameter (categorical variable) in the logistic regression model, but the model output listed them separately. Bedrock unit OZc was defined as reference, so it's not shown in the output.

Table S2. Documented bedrock aquifers with elevated arsenic of natural source

Country/region	Area (km^2) (number of analysis)	Arsenic concentration range, median and/or mean (μg L^{-1}), and exceedance rate	Aquifer rock type	Groundwater conditions, arsenic distribution and its associations	Reference
Northern Bavaria, Germany	8,000 (n=424)	maximum [As] = 150 μg L^{-1} 37% >10 μg L^{-1}	Upper Triassic interbedded mudstone and sandstone of regressive-continental character	pH 6.7–7.8 EC 400–700 μS cm^{-1} Mn (~0.5 mg L^{-1}), Fe (~0.5 mg L^{-1})	Heinrichs and Udluft (1999)
Obuasi and Bolgatanga, Ghana	1,600 and 900, respectively	<0.4 - 64 μg L^{-1}	Proterozoic meta-sedimentary rocks of Birimian formation (schist, phyllite and greywache with minor tuff) and meta-volcanic rock (greenstone, amphibolites and tuff with minor schist and phyllite), with granitic intrusions	pH 3.9-6.8 (Obuasi) pH 6.0-7.4 (Bolgatanga) groundwater [As] associated with higher pH and HCO_3, highest [As] at depth of 40-70m, then lower in deeper because reducing condition limited As oxidation and release	Smedley (1996)
Ankobra Basin, Ghana	8,400 (n=64)	<0.05 – 491 μg L^{-1} median 0.05 μg L^{-1} mean 6.5 μg L^{-1} 5% >10 μg L^{-1}	lower Proterozoic rocks, including 60% of Lower Birimian (mainly pelitic origin and consisting of great thicknesses of alternating shales, phyllites, greywacke and argillaceous beds with tuffs and lavas) and 8% of Upper Birimian (volcanic and pyroclastic origin), overlaid by 15% of Tarkwaian system consisting of thick clastic sequence, intruded by 10% of granitoid ranging from felsite and quartz porphyry to meta-dolerite, gabbro and norite	pH 3.89–6.78 (mean 5.70 and median 5.72) EC median 230 μS cm^{-1} HCO_3 dominant type, no cations dominant high Fe and Mn groundwater undersaturated with major carbonate species	Kortatsi (2007)
Southern Ghana	~50,000 (n=286)	5-12% >10 μg L^{-1}	Birimian formation (meta-volcanics) and Tarkwaian formation (meta-sediments of conglomerates, sandstones, quartzite and shale), with intrusive granitoids Voltaian formation (Paleozoic consolidated sedimentary rocks of sandstones, shales, arkoses, mudstones, sandy, limestone, etc.)	high Fe and Mn highest [As] mostly in Birimian formation	Buamah et al. (2008)

Northern Burkina Faso	~4,500 (n=36)	<0.5-1630 μg L^{-1} median 15.1 μg L^{-1} mostly As(V) (earlier regional research showing 13% >10 μg L^{-1})	high [As] mostly in Birimian volcano-sedimentary schists	pH 5.77-7.78 (median 7.14) median DO 0.75mg L^{-1} Eh mostly around 300+ mV NO_3 median 1.05 mg L^{-1} low Cl (mostly below 6 mg L^{-1}) water type Ca(-Mg-Na)-HCO_3 high As often found in deeper wells, negatively correlated with DO, positively correlated with Mo	Smedley et al (2007)
Chattisgarh, central India	~120 (n=91)	<1-250 μg L^{-1}	max [As] = 13 μg L^{-1} in sandstone, shale, and limestone; max [As] = 15 μg L^{-1} in granite; max [As] = 250 μg L^{-1} in granite with pegmatite, basalt, and dolerite	pH 6.4-8.8 EC 200-6000 μS cm^{-1}	Shukla et al (2010)
Zimapan Valley, Mexico	800 (n=54)	<14-1000 μg L^{-1} 52% >14 μg L^{-1} in volcanic rock 19-43 μg L^{-1} 22% >14 μg L^{-1}	fractured Cretaceous thin-bedded shaly limestone interbedded with laminated calcareous shales, Tertiary volcanic rocks, intensely folded and faulted during Paleocene–Lower Oligocene Hidalgoan or Laramide Orogeny	pH 5.83-8.48 (mostly 6.9-8.1) Eh 147-572 mV (mostly 360-500) highest [As] in low Eh water	Armienta et al (2001)
Bowen Island, British Columbia, Canada	~ 2 (n=26)	0.5-580 μg L^{-1}	the Bowen Island Group (mafic to intermediate volcanic flows, interbedded with volcaniclastic sandstone, siliceous argillite, chert and tuff, metamorphosed to greenschist), intruded by Middle to Late Jurassic granodiorite to quartz monzonite	pH 7.40-8.86 EC 113-1033 μS cm^{-1} Ca-Na-K-HCO_3 type water	Boyle et al (1998)
Southeastern Michigan	~15,000 (n=73)	0.5-278 μg L^{-1} mean 29 μg L^{-1} median 16 μg L^{-1} 68% >10 μg L^{-1} mostly As(III)	the Paleozoic-aged Coldwater Shale (gray to dark gray shale), carbonate (limestone or dolomite, particularly in the western half of the basin), siltstone, and sandstone, interbedded with Marshall sandstone	pH 6.9-8.5 (mean=median=7.4) alkalinity 185-628 mg L^{-1} Ca(Na)-HCO_3 type As negative related with DO high As associated with Coldwater shale and Marshall sandstone, as well as overlying glacial drift	Kim et al (2002) Meliker et al (2009)
Orange County, North Carolina	~700 (n=471)	<1 - 8 μg L^{-1} 70% < 1 μg L^{-1}	Plutons and intrusive; felsic and mafic lavas and tuffs; welded tuffs and hydrothermal quartz bodies; and air fall tuffs and epiclastic rocks	Wells deeper and closer to transition zone, faults and plutons more likely show high As.	Kim et al (2011)

Newark Basin, Pennsylvania	~1,000 (n=53)	0.01-65 μg L^{-1} median 2.85 μg L^{-1} 23% >10 μg L^{-1} mostly As(V)	the Triassic Passaic Formation (quartzose sandstone grading into red mudstone, with some interbedded gray shale and argillite) and Lockatong deposits (dark siltstones, shales, argillites); Cambrian & Precambrian quartzite and gneiss; Jurassic diabase	high groundwater [As] in Lockatong and Passaic formation high As in groundwater with neutral to high pH, low NO_3 and Cl^-	Peters and Burkert (2008)
New England Coastal Basin	~60,000 (n=804+58)	20% >5 μg L^{-1} 11% >10 μg L^{-1}	Mc (primarily metasedimentary calcareous or calc-silicate rocks) with 44% [As] >5 μg L^{-1}, Ms (metasedimentary group) with 4% [As] >5 μg L^{-1}, Mu (undifferentiated metamorphosed marine sediments) with 27% [As] >5 μg L^{-1}; If (mostly felsic igneous rocks, primarily granites) with 12% [As] >5 μg L^{-1}, and other igneous rocks	arsenic most common in groundwater with high pH	Ayotte et al (1999, 2003)
New England	186,460 (n=2,470)	23% > 5 μg L^{-1}	as above	bedrock geology, lithochemistry, stream sediment [As], Pleistocene marine inundation and intrusive granitic pluton are among the most significant factors controlling groundwater [As]	Ayotte et al (2006)
Eastern Highlands of Connecticut	~400 (n=225)	maximum [As]= 83 μg L^{-1} 6% >5 μg L^{-1} 2% >10 μg L^{-1}	Precambrian and Paleozoic Brimfield schist (interlayered dark-gray schist and greenish gray, fine- to medium-grained calc-silicate gneiss), and the Hebron Formation (gray, rusty weathering, medium- to coarse-grained, interlayered schist and gneiss), frequently intruded by granitic and pegmatitic sills and dikes	7/13 of high groundwater As in Hebron Formation	Pagach (2009)
East-central Massachusetts	~8,000 (n=478)	<0.2 - 1540 μg L^{-1} 13% >10 μg L^{-1}	Devonian-Silurian meta-sedimentary rocks and Devonian granite-diorite intrusions, crossed by major faults	Statistically significant differences among distributions by bedrock unit for arsenic	Colman (2011)
Northern Vermont	800 (n=45)	<1 - 327 μg L^{-1} 27% >10 μg L^{-1} mean 16.5 μg L^{-1}	Ordovician phyllite (metamorphosed shales and sandstones), greenstone (metabasalt) and isolated pods of serpentinite and associated ultramafic rocks, covered by glacial tills	elevated As occurrences have a general spatial association with ultramafic rock bodies; high As may be enhanced by alkaline pH and high HCO_3^-	Ryan et al (2011)

Southeastern New Hampshire	5,300 (n=353)	14-21% (overall 19%) >10 µg L^{-1}	high groundwater [As] related with bedrock geology		Montgomery et al (2003)
South central New Hampshire	225 (n=127)	2-400 µg L^{-1} median 16 µg L^{-1} 62% >10 µg L^{-1}	Silurian metapelite intruded by late Devonian granite (grading locally to tonalite) and pegmatites, with a variably thin cover of glacial till and outwash	high As groundwater samples have low Fe, distributed mainly in pegmatite zone	Peters and Blum (2003)
New Hampshire	24,217 (n=218)	~15% >10 µg L^{-1}	high As clustered in metasedimentary rocks of the upper amphibolite to granulite metamorphic facies, plutons of Devonian granite, and in metasedimentary rocks of the lower amphibolite metamorphic facies and plutons of Devonian granodiorites		Peters et al (1999)
Buxton and Hollis, Maine	~200 (n=1,111)	<5 - >300 µg L^{-1} 55% >5 µg L^{-1} 14% >50 µg L^{-1}	Silurian-Devonian meta-sedimentary schist (sandstone, shale, limestone) intruded by granitic rocks	higher [As] groundwater in meta-sedimentary rock units than in granitic rocks	Marvinney et al (1994)
Goose River Basin, Maine	~30 (n=36 in 2001, n=96 in 2002 and 2003)	<1 – 46 µg L^{-1} 28% >10 µg L^{-1}in 2001 36% > 10 µg L^{-1} in 2003 mostly As(V)	Bucksport Formation (amphibolite-grade metasediments with pyritic schistose, with median whole rock As of 40 mg kg^{-1}); Waldoboro garnet-bearing granitoid (strongly sheared anatectic granitoids and late pegmatitic phases, with median whole rock As of 46 mg kg^{-1}); Waldoboro quartz diorite; Waldoboro granite porphyry	DO 4.1-6.9 mg L^{-1} high groundwater [As] in Bucksport formation and Waldoboro garnet-bearing granitoid, associated with high $^{18}O_{so4}$	Sidle et al (2001, 2002, 2003)
Northport, Maine	~2 (n=35)	69% >10 µg L^{-1} mean 270 µg L^{-1} mostly As(III)	fractured-bedrock system composed of sulfidic schist with granitic to dioritic intrusions	pH 5.8-8.5 high As associated with high pH, low Eh, and high Fe Ca-HCO_3 type water having low [As], Na-HCO_3 type high [As]	Lipfert et al (2006)
Greater Augusta, Maine	1,135 (n=790)	<0.07 – 325 µg L^{-1} mean 12.2 µg L^{-1} median 3.8 µg L^{-1} 31% > 10 µg L^{-1}	Silurian meta-sedimentary rocks intruded by Devonian granitic rocks, Ordovician-Cambrian volcanic rocks	pH median 7.59 DO median 1.5 mg L^{-1} alkalinity median 1.28 mmol/L highest groundwater [As] in meta-sedimentary rocks, followed by granite intrusions, and lowest in old volcanic rocks	Yang et al (2009) This research

Reference list for Table S2

(1) Heinrichs, G.; Udluft, P., Natural arsenic in Triassic rocks: A source of drinking-water contamination in Bavaria, Germany. *Hydrogeology Journal* **1999,** *7*, (5), 468-476.
(2) Smedley, P. L., Arsenic in rural groundwater in Ghana. *Journal of African Earth Sciences* **1996,** *22*, (4), 459-470.
(3) Kortatsi, B., Hydrochemical framework of groundwater in the Ankobra Basin, Ghana. *Aquatic Geochemistry* **2007,** *13*, (1), 41-74.
(4) Buamah, R.; Petrusevski, B.; Schippers, J. C., Presence of arsenic, iron and manganese in groundwater within the gold-belt zone of Ghana. *Journal of Water Supply: Research & Technology-AQUA* **2008,** *57*, (7), 519-529.
(5) Smedley, P. L.; Knudsen, J.; Maiga, D., Arsenic in groundwater from mineralised Proterozoic basement rocks of Burkina Faso. *Applied Geochemistry* **2007,** *22*, 1074-1092.
(6) Shukla, D. P.; Dubey, C. S.; Singh, N. P.; Tajbakhsh, M.; Chaudhry, M., Sources and controls of Arsenic contamination in groundwater of Rajnandgaon and Kanker District, Chattisgarh Central India. Journal of Hydrology **2010**, 395, 49-66.
(7) Armienta, M. A.; Villasenor, G.; Rodriguez, R.; Ongley, L. K.; Mango, H., The role of arsenic-bearing rocks in groundwater pollution at Zimapan Valley, Mexico. *Environmental Geology* **2001,** *40*, (4-5), 571-581.
(8) Boyle, D. R.; Turner, R. J. W.; Hall, G. E. M., Anomalous arsenic concentrations in groundwaters of an island community, Bowen Island, British Columbia. *Environmental Geochemistry and Health* **1998,** *20*, (4), 199-212.
(9) Kim, M.-J.; Nriagu, J.; Haack, S., Arsenic species and chemistry in groundwater of southeast Michigan. *Environmental Pollution* **2002,** *120*, (2), 379-390.
(10) Meliker, J. R.; Slotnick, M. J.; AvRuskin, G. A.; Haack, S. K.; Nriagu, J. O., Influence of groundwater recharge and well characteristics on dissolved arsenic concentrations in southeastern Mihchigan groundwater *Environmental Geochemistry and Health* **2009,** *31*, 147-157.
(11) Kim, D.; Miranda, M. L.; Tootoo, J.; Bradley, P.; Gelfand, A. E., Spatial Modeling for Groundwater Arsenic Levels in North Carolina. Environmental Science & Technology **2011**, 45, (11), 4824-4831.
(12) Peters, S. C.; Burkert, L., The occurrence and geochemistry of arsenic in groundwaters of the Newark basin of Pennsylvania. *Applied Geochemistry* **2008,** *23*, (1), 85-98.
(13) Ayotte, J. D.; Nielsen, M. G.; Robinson, G. R.; Moore, R. B., Relation of Arsenic, Iron, and Manganese in Ground Water to Aquifer Type, Bedrock Lithogeochemistry, and Land Use in the New England Coastal Basins. *U.S. Geological Survey, Water-Resources Investigations Report 99-4162.* **1999**, P70.
(14) Ayotte, J. D.; Montgomery, D. L.; Flanagan, S. M.; Robinson, K. W., Arsenic in groundwater in eastern New England: occurrence, controls, and human health implications. *Environmental Science and Technology* **2003,** *37*, (10), 2075-2083.
(15) Ayotte, J. D.; Nolan, B. T.; Nuckols, J. R.; Cantor, K. P.; Robinson, G. R.; Baris, D.; Hayes, L.; Karagas, M.; Bress, W.; Silverman, D. T.; Lubin, J. H., Modeling the Probability of Arsenic in Groundwater in New England as a Tool for Exposure Assessment. *Environ. Sci. Technol.* **2006,** *40*, (11), 3578-3585.

(16) Pagach, J. J., Distribution of Arsenic in Bedrock Wells in the Eastern Highlands of Connecticut. Report in *New England Private Well Sysposium, 2009, Portland, ME.* **2009**.
(17) Colman, J. A., Arsenic and uranium in water from private wells completed in bedrock of east-central Massachusetts—Concentrations, correlations with bedrock units, and estimated probability maps. *U.S. Geological Survey Scientific Investigations Report* **2011**, 5013, 113p.
(18) Ryan, P. C.; Kim, J.; Wall, A. J.; Moen, J. C.; Corenthal, L. G.; Chow, D. R.; Sullivan, C. M.; Bright, K. S., Ultramafic-derived arsenic in a fractured bedrock aquifer. *Applied Geochemistry* **2011**, 26, (4), 444-457.
(19) Montgomery, D. L.; Ayotte, J. D.; Carroll, P. R.; Hamlin, P., Arsenic concentrations in private bedrock wells in southeastern New Hampshire, *U.S. Geological Survey Fact Sheet 051-03*. **2003**.
(20) Peters, S. C.; Blum, J. D., The source and transport of arsenic in a bedrock aquifer, New Hampshire, USA. *Applied Geochemistry* **2003,** *18*, (11), 1773-1787.
(21) Peters, S. C.; Blum, J. D.; Klaue, B.; Karagas, M. R., Arsenic Occurrence in New Hampshire Drinking Water. *Environ. Sci. Technol.* **1999,** *33*, (9), 1328-1333.
(22) Marvinney, R. G.; Loiselle, M. C.; Hopeck, J. T.; Braley, D.; Krueger, J. A. Arsenic in Maine ground water - An example from Buxton, Maine, *The 1994 Focus Conference on Eastern Regional Ground Water Issues, Burlington, Vermont*, **1994**, pp 701-715.
(23) Sidle, W. C.; Wotten, B.; Murphy, E., Provenance of geogenic arsenic in the Goose River basin, Maine, USA. *Environmental Geology* **2001,** *41*, (1-2), 62-73.
(24) Sidle, W., 18OSO4 and 18OH2O as prospective indicators of elevated arsenic in the Goose River ground-watershed, Maine. *Environmental Geology* **2002,** *42*, (4), 350-359.
(25) Sidle, W. C.; Fischer, R. A., Detection of H-3 and Kr-85 in groundwater from arsenic-bearing crystalline bedrock of the Goose River basin, Maine. *Environmental Geology* **2003,** *44*, (7), 781-789.
(26) Lipfert, G.; Reeve, A. S.; Sidle, W. C.; Marvinney, R., Geochemical patterns of arsenic-enriched ground water in fractured, crystalline bedrock, Northport, Maine, USA. *Applied Geochemistry* **2006,** *21*, (3), 528-545.
(27) Yang, Q.; Jung, H. B.; Culbertson, C. W.; Marvinney, R. G.; Loiselle, M. C.; Locke, D. B.; Cheek, H.; Thibodeau, H.; Zheng, Y., Spatial Pattern of Groundwater Arsenic Occurrence and Association with Bedrock Geology in Greater Augusta, Maine. *Environmental Science & Technology* **2009,** *43*, (8), 2714-2719.