# Arsenic in Groundwater in Eastern New England: Occurrence, Controls, and Human Health Implications

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In eastern New England, high concentrations (greater than 10 $\mu$g/L) of arsenic occur in groundwater. Privately supplied drinking water from bedrock aquifers often has arsenic concentrations at levels of concern to human health, whereas drinking water from unconsolidated aquifers is least affected by arsenic contamination. Water from wells in metasedimentary bedrock units, primarily in Maine and New Hampshire, has the highest arsenic concentrations—nearly 30% of wells in these aquifers produce water with arsenic concentrations greater than 10 $\mu$g/L. Arsenic was also found at concentrations of 3–40 mg/kg in whole rock samples in these formations, suggesting a possible geologic source. Arsenic is most common in groundwater with high pH. High pH is related to groundwater age and possibly the presence of calcite in bedrock. Ion exchange in areas formerly inundated by seawater also may increase pH. Wells sampled twice during periods of 1–10 months have similar arsenic concentrations (slope = 0.89; r-squared = 0.97). On the basis of water-use information for the aquifers studied, about 103 000 people with private wells could have water supplies with arsenic at levels of concern (greater than 10 $\mu$g/L) for human health.

## Introduction

Arsenic is a common trace element in groundwater and is present in varying amounts across the United States (*1, 2*). The presence of arsenic in drinking water has been associated with adverse health outcomes, primarily cancers, and currently is regulated by Federal and State standards for public water systems (*3*). The final ruling by the U.S. Environmental Protection Agency (USEPA) on arsenic in drinking water lowered the standard from 50 $\mu$g/L to 10 $\mu$g/L (*4*). In New England, moderate to high (10–50 $\mu$g/L) concentrations of arsenic are known to occur in groundwater, but the occurrence and distribution has not previously been quantified at the regional scale. Data from randomly selected private domestic wells throughout Maine show that only about 1–3% of the wells have water with arsenic concentrations above 50 $\mu$g/L, but that more than 12% of wells have concentrations that exceed 10 $\mu$g/L (*5*). In a cancer epidemiology study in New Hampshire, data from private and public wells used by 99 control subjects showed that approximately 5% of the wells had water with arsenic concentrations above 50 $\mu$g/L (*6*).

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Previous studies on arsenic in groundwater in the region do not identify an arsenic source (*7*). Increasing evidence shows that the source of the arsenic in New England is dominantly natural and originates from minerals in the rocks of the region (*2, 8–10*). Studies in other parts of the United States and Canada also have related arsenic to geologic sources (*11–13*). Anthropogenic sources of arsenic (such as former pesticide use, treated lumber, and manufacturing) also may contribute to groundwater contamination, but studies from this region are inconclusive (*14, 15*).

The natural chemistry of groundwater is related to the geochemistry of the aquifer materials through which it flows and to the residence time of the water in those aquifer materials (*16, 17*). On the basis of past studies in New England, water chemistry is expected to depend partly on the geochemical character of the underlying bedrock formations and partly on the degree of metamorphism of the bedrock (*16, 18*). Unconsolidated aquifers in the study area generally contain recently recharged groundwater and have similar major ion water chemistry. Water quality in bedrock aquifers in eastern New England, however, is highly variable, and the chemistry of the water can be markedly different from well to well; the most common groundwater contaminants include iron and manganese, arsenic, and radionuclides.

Water from public-supply systems is mostly withdrawn from unconsolidated aquifers and accounts for about 30% of all drinking water in New England (*19*). Conversely, privately supplied water is mostly withdrawn from bedrock aquifers and accounts for about 20% of all drinking-water supplies in New England. (privately supplied groundwater accounts for 40% of the total drinking-water supply in northern New England (*20*) and comes primarily from bedrock aquifers, as compared to less than 15% in southern New England). The remaining 50% of the water supply for New England is from surface-water sources (*19*).

This study characterizes the regional occurrence and distribution of arsenic in unconsolidated and bedrock aquifers in parts of Maine, New Hampshire, eastern Massachusetts, and Rhode Island (Figures 1 and 2). Current monitoring data, in conjunction with extensive existing data from Safe Drinking Water Act compliance monitoring, are used. This study is part of the U.S. Geological Survey (USGS) National Water-Quality Assessment (NAWQA) Program. The study area was determined by national and local program goals, and it covers some of the most populated parts of New England, coinciding with areas where large amounts of groundwater are used for water supply. Other goals of this study were to identify region-scale controls on occurrence and mobility of arsenic and to identify human health implications by estimating the size of the population using high-arsenic water supplies. Water samples were collected from randomly selected public and private wells in the region that are currently used for domestic drinking-water supply. The wells are completed in the two principal aquifer types that underlie the study area—(1) unconsolidated aquifers, primarily composed of glacial stratified sand and gravel, which are discontinuous and mostly are confined to river-valley settings, and (2) fractured crystalline-bedrock aquifers, which are continuous and underlie the entire study area (*21*). Bedrock aquifers can be further characterized in terms of the general geochemical composition, according to how they might influence the chemistry of water that flows through them (*18, 22, 23*). Bedrock unit descriptions from state geologic maps were used to categorize the aquifers; metamorphosed marine sediments described as "variably calcareous" (5–50% calcite) or derived from calcareous pro-
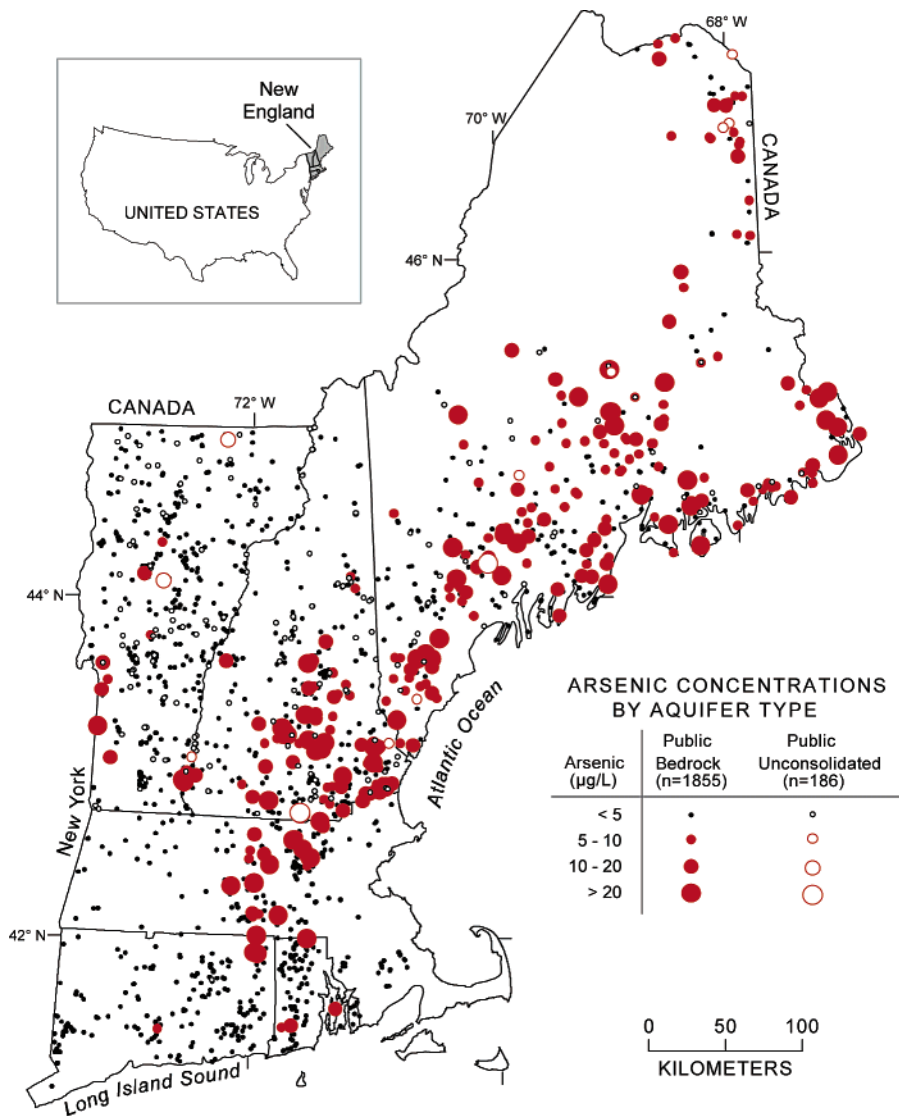
FIGURE 1. Arsenic concentration in source waters to public-supply wells in New England.

toliths were grouped in the category $M_c$; undifferentiated metamorphosed marine sediments in the $M_u$ group and felsic igneous rocks, such as granites, in the $I_f$ group.

## Study Design and Methods

The current study design is based on a recent retrospective study of over 800 wells, used for public supply (7). The arsenic data in the retrospective study were collected to represent aquifer conditions as much as possible, by selecting single well water systems that have little or no treatment before delivery. In addition, we expanded the scope of the original retrospective study to include an additional 800 public supply wells across all of New England (Figure 1). These data clearly indicate that the extent of significant arsenic contamination in wells is in the eastern part of New England. The current study addresses arsenic in groundwater in the eastern part of New England because of the study unit design of the USGS's NAWQA program and the apparent focus of the arsenic problem.

This study also builds on relations found in the retrospective study to aid in the design of the well networks used to characterize arsenic occurrence. Relations were identified between arsenic and aquifer type, and, in the bedrock aquifer, to specific groups of geologic formations; these groups are described in brief below and in detail in the retrospective

study (7). In eastern New England, arsenic concentrations were significantly higher in water from wells drilled into bedrock aquifers than in water from wells in unconsolidated aquifers. Further, arsenic concentrations were also greater in metamorphosed marine sediments ($M_c$) described as variably calcareous or derived from calcareous protoliths on State geologic maps, than in other metasedimentary ($M_u$) and felsic igneous ($I_f$) rocks (7).

The current study compares arsenic concentrations in water from unconsolidated and bedrock aquifers and from wells in different types of bedrock. Three sampling networks of approximately 30 wells each, based on models of aquifer type and generalized geologic information, were used (7, 18, 23). Thirty wells are in unconsolidated aquifers, and 58 wells are in bedrock aquifers (Figure 2). While this number of wells is small for regional characterization of arsenic, the incorporation of the retrospective study data and careful stratification and random selection of sampling sites during study design provides adequate data for occurrence and distribution analysis. All wells are selected from databases of georeferenced public and private wells in conjunction with an equal-area grid stratified random-selection routine combined with a geographic information system (GIS) (24). All 88 wells were sampled between September 1999 and October 2001.
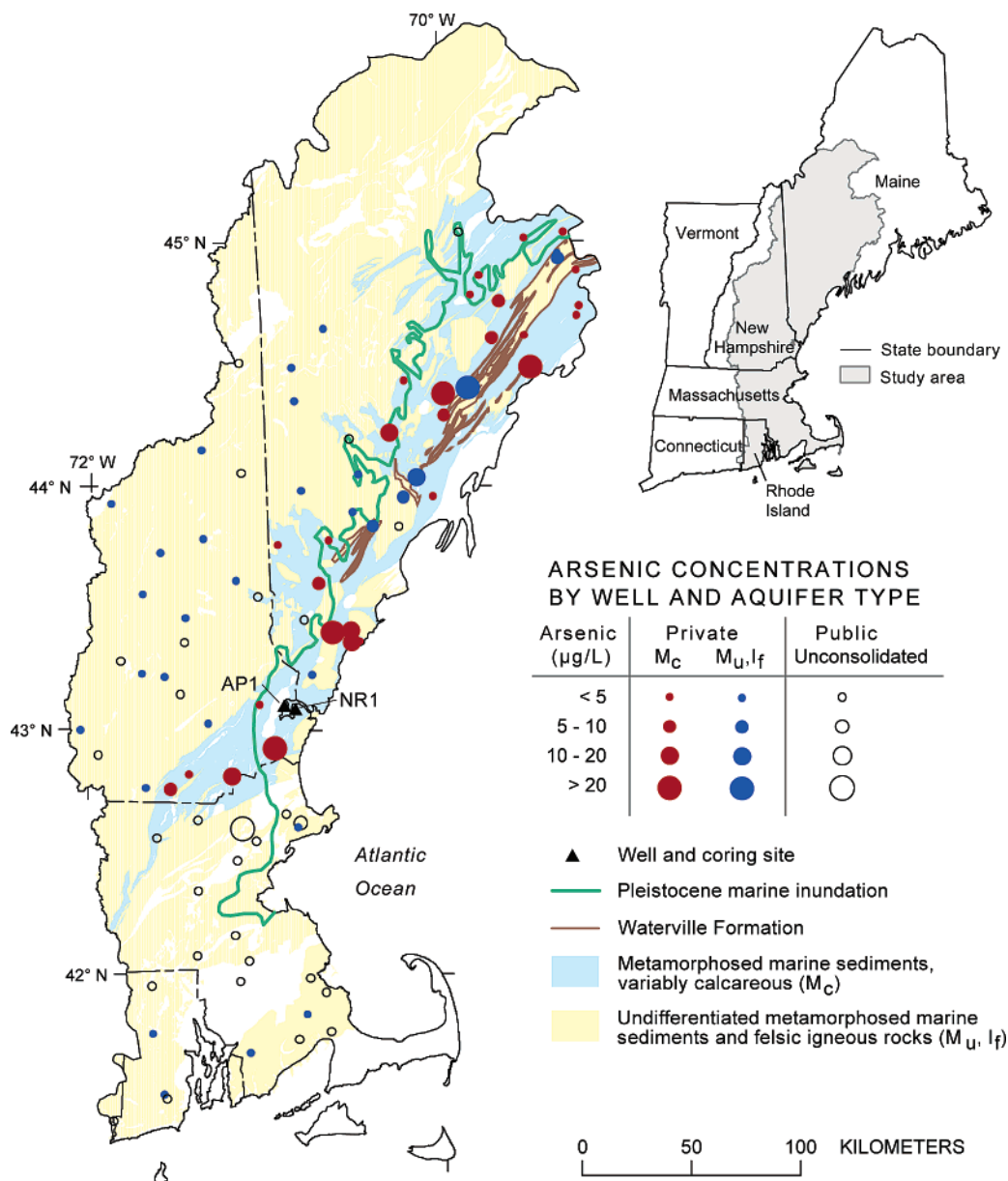
**FIGURE 2.** Arsenic concentrations in water from private, domestic, bedrock wells, and public, unconsolidated wells in eastern New England.

In an associated small site-specific study in the area, two bedrock wells were drilled and bedrock core (Figure 2) was collected so that solid-phase and aqueous geochemistry could be analyzed. Water from individual fractures in the wells was sampled by isolating discrete intervals in the boreholes, using inflatable packer technology. Groundwater arsenic concentrations from discrete fractures were compared to whole rock arsenic concentrations from the same fracture zone.

**Wells in Unconsolidated Aquifers.** The network of wells in the unconsolidated aquifers is from a database of public-supply wells compiled from each of the states in the study area. The 30 wells in the unconsolidated aquifers are public-water-supply wells, screened in sand and gravel, that are currently (2002) in use. These wells range in depth from 8 to 50 m, have a median depth of 16 m, and have a median yield of 1117 L/min.

**Wells in Bedrock Aquifers.** Sets of wells were sampled in two general bedrock groups (25). The first sampling network consists of 28 bedrock wells in the lithochemical group composed mainly of metamorphosed marine sediments described as variably calcareous ($M_c$). These rocks form a band that runs roughly parallel to the regional bedrock strike (approximately north-northeast), from central Massachusetts through southeastern New Hampshire and south-central Maine (Figure 2). The second sampling network consists of 30 bedrock wells in a combination of undifferentiated metamorphosed marine sediments ($M_u$) and felsic igneous ($I_f$) rocks (Figure 2). Together, these bedrock units cover more than 85% of the study area.

All 58 bedrock wells are privately owned and used for domestic supply, are less than 20 years old, and are generally constructed to similar specifications, using 0.15−0.2 m-diameter steel casing. The median depth of the wells is 80 m, median casing length is 9 m, and the median reported yield is 19 L/min. Casings are typically installed through the unconsolidated glacial sediments and driven into the underlying bedrock. Groundwater enters the wells through fractures that intersect the open borehole.

**Sampling and Analysis.** Water samples were collected and analyzed for major ions, trace elements, chlorofluoro-carbons (CFCs), and other constituents (see Supporting Information), in accordance with NAWQA protocols (26−

**TABLE 1. Percentage of Wells with Arsenic Concentrations Exceeding 5, 10, 20, and 50 $\mu$g/L, by Aquifer Type[a]**

| aquifer type | N | percentage of wells with arsenic concentrations exceeding: ($\mu$g/L) | | | |
|---|---|---|---|---|---|
| | | 5 | 10 | 20 | 50 |
| *Well Sampled for This Study* | | | | | |
| unconsolidated | 30 | 7 | 3 | 3 | 0 |
| crystalline bedrock | 58 | 31 | 17 | 9 | 2 |
| $M_c$ | 28 | 46 | 29 | 14 | 4 |
| $M_u$, $I_f$ | 30 | 17 | 7 | 3 | 0 |
| *Wells from Retrospective Study (7)* | | | | | |
| unconsolidated | 145 | 7 | 3 | 3 | <1 |
| crystalline bedrock | 645 | 20 | 11 | 5 | 1 |
| $M_c$ | 187 | 41 | 20 | 8 | 2 |
| $M_u$, $I_f$ | 458 | 14 | 7 | 3 | 1 |

*[a] N, number of wells; <, less than.*

*28*). During the initial reconnaissance visit, an unfiltered sample was collected and analyzed for total arsenic (*27*). For wells that met NAWQA criteria, a second visit was made; before collection of any samples, wells were pumped until water temperature, dissolved oxygen, specific conductance, and pH were stable (*27*). Cleaned fluorinated ethylene polypropylene (FEP) tubing with stainless steel fittings was connected to the water line just ahead of the pressure tank, distribution lines, and any treatment devices such as ion-exchange filters. Samples were collected at a flow rate of 0.05 L/min in a portable sampling chamber. Samples for arsenic and other trace elements were collected through a 0.45-$\mu$m disposable capsule filter and were acidified with ultrapure nitric acid to a pH of less than 2. The analyses for total arsenic and other trace elements were made at the USGS National Water Quality Laboratory (NWQL) and at the State of New Hampshire, Department of Environmental Services Laboratory, using ICP/MS methods with a laboratory reporting level (LRL) of 0.9–2 $\mu$g/L (*29*). All analyses of arsenic are corrected for physical and spectral interferences (*30*) according to standard USGS methods (*29*).

Quality-control (QC) samples, including replicates and field blanks, were prepared according to NAWQA protocols (*27*). Fifteen blanks (17%) and seven replicates (8%) were collected for trace elements for the 88 wells. Analysis of all blank samples showed no inherent bias in the method of analysis for the trace elements and major ions of interest. Replicate samples were collected immediately after the routine samples in the field using the same collection methods and equipment. All differences measured in concentrations between replicate pairs were well within the precision of the method for all trace elements.

## Results and Discussion

Concentrations of arsenic above the current USEPA drinking water standard of 10 $\mu$g/L were almost entirely from bedrock aquifers. Bearing this out, only one unconsolidated aquifer well in the 30 wells sampled (3%) had a concentration of arsenic in water exceeding 10 $\mu$g/L, whereas 17% of the private bedrock wells sampled had water with arsenic concentrations exceeding 10 $\mu$g/L (Table 1). These results are similar to, or slightly higher than, those reported in the associated retrospective study (*7*) that analyzed data from public wells in both unconsolidated and bedrock aquifers (Table 1). In recent statewide studies, 12–13% of bedrock wells in Maine have arsenic concentrations exceeding 10 $\mu$g/L (*5*); in New Hampshire, 2% of public supply wells and 13% of private domestic wells have arsenic exceeding 10 $\mu$g/L (*9*).

**Relation of Arsenic Concentrations to Regional Geology.** The results of the analyses of water samples from the 58

bedrock wells were evaluated for statistical differences based on regional geology and other factors. The nonparametric Mann–Whitney test (*31*) indicated arsenic concentrations in water from bedrock wells in the variably calcareous metasedimentary rocks ($M_c$) were significantly higher ($p = 0.001$) than in water from bedrock wells in the combined undifferentiated metasedimentary and felsic igneous groups ($M_u$ and $I_f$).

Nearly half (46%) of the wells sampled in the $M_c$ group have water with arsenic concentrations greater than 5 $\mu$g/L, and 29% were greater than 10 $\mu$g/L (Table 1). In the $M_u$ and $I_f$ group, only about 17% of wells have water with arsenic concentrations greater than 5 $\mu$g/L and 7% were greater than 10 $\mu$g/L (Table 1, Figure 2).

The location of the wells in the $M_u$ and $I_f$ group that have measurable concentrations of arsenic is important to consider. In general, water from wells in the $M_u$ and $I_f$ group has little or no arsenic (<1 $\mu$g/L), with the exception of water from wells in the Waterville Formation in Maine. The Waterville Formation (*32*), although geographically near the rocks of the $M_c$ group, is not described as calcareous on the State geologic map and thus was included in the $M_u$ and $I_f$ group. Water in four of the five private wells in the $M_u$ and $I_f$ group (in the Waterville Formation) had arsenic concentrations ranging from 5.7 to 48 $\mu$g/L (Figure 2). Similarly, in the retrospective study, 8 of the 10 public-supply bedrock wells in the Waterville Formation had arsenic concentrations exceeding 5 $\mu$g/L (*7*).

To investigate the possible source of arsenic, continuous rock core was collected during the installation of two 30-m test wells in bedrock formations of the $M_c$ group (Figure 2). Both wells yielded core with visible sulfide mineralization associated with thin layers of calcite in the bulk rock and on water-bearing fractures (*33*). Arsenic was present in the sulfide minerals (pyrite and pyrrhotite with accessory chalcopyrite) associated with the calcite in the cores (*34*). Arsenic concentrations in the water samples are hypothesized to be related to solid-phase arsenic concentrations. Arsenic concentrations in whole-rock samples near water bearing fracture intervals from well and core sites NR-1 and AP-1 averaged 15 mg/kg and ranged from 3 to 40 mg/kg (*35*). Water samples collected at the same fracture intervals as the whole rock samples have arsenic concentrations ranging from 0.74 to 6.1 $\mu$g/L. Data from rocks in this study are on the upper end of the average crustal arsenic concentrations in igneous rocks, limestone, sandstone, and shale, which have been reported as 1.5, 2.6, 4.1, and 14.5 mg/kg, respectively (*36*). In New Hampshire, a geologic model suggests that late-stage pegmatites, formed during granite crystallization, can be enriched with arsenic and provide a source of arsenic to groundwater for specific bedrock formations (*9*).

**Arsenic Variation.** The variation of arsenic concentrations in groundwater is an important consideration for monitoring, regulation, and treatment of arsenic as well as for environmental and health studies concerned with arsenic. Variation in arsenic concentrations is related to the following general factors: (1) natural variation in the groundwater system, (2) affect of anthropogenic factors, and (3) laboratory analytical variability (*12*). With the data collected for this study, it was possible to evaluate only the apparent variability in arsenic concentrations in water from unfiltered samples collected during field reconnaissance at 43 of the 58 bedrock wells and arsenic concentrations for the filtered samples collected between 1 and 12 months later. The absolute value of intrawell differences ranged from 0.024 to 12.65 $\mu$g/L, with a median difference of 0.41 $\mu$g/L (Figure 3). Only four of the 43 wells had a difference in arsenic concentration greater than 2 $\mu$g/L. Concentrations of arsenic from the two sets of samples were highly correlated ($r^2 = 0.97$, $p < 0.001$); a linear regression indicates that the unfiltered samples are about 11% higher
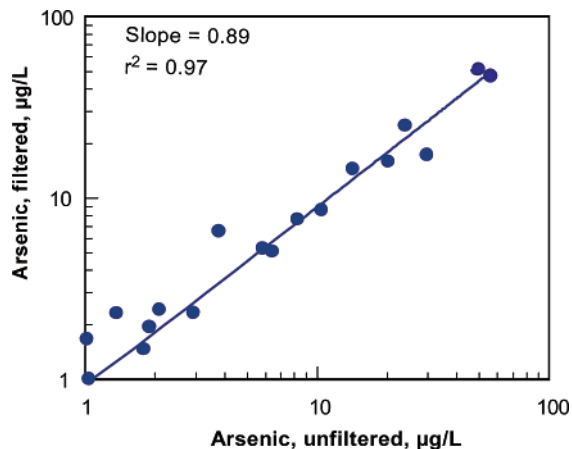
FIGURE 3. Relation between arsenic concentrations from filtered and unfiltered samples.
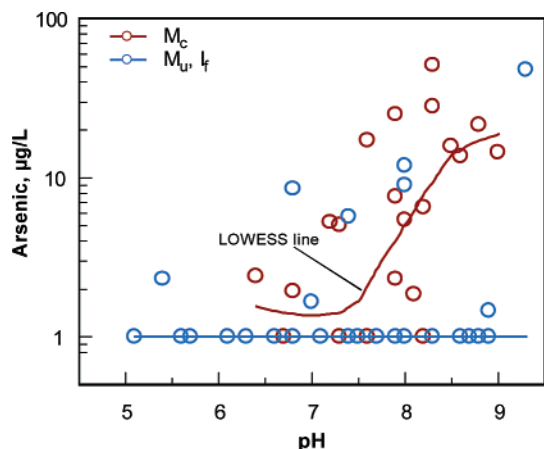


FIGURE 4. Relation between pH values and arsenic concentrations by aquifer type.



FIGURE 5. Relation between age of water and (a) pH and (b) arsenic concentrations.

than the filtered samples (slope = 0.89). These results suggest that arsenic in water from the private domestic bedrock wells is primarily dissolved but that a small fraction may be particulate. Variation in arsenic concentrations is likely due to sampling protocol differences between filtered and unfiltered samples; however, natural variation or laboratory variation cannot be dismissed. Data presented in a biomarker study in New Hampshire (6) that considered arsenic variability in groundwater found little variation in two arsenic samples collected between 3 and 5 years apart from domestic wells.

**Chemical and Physical Controls.** In the groundwater sampled for this study, high arsenic groundwater ($>10 \mu g/L$) generally has pH values of 8 or greater (Figure 4). This broad relation between arsenic and pH has been noted for groundwater across the United States (2) and elsewhere (37, 38) and is a control for sorption/desorption processes. Laboratory studies demonstrate that arsenate [As(V)] desorption increases at progressively higher pH on a variety of solids, including iron oxides and some common clay minerals (39, 40). Arsenite [As(III)] desorption also is pH dependent, although higher pH conditions are required for desorption from some of these same solids (39). Another factor that can affect arsenic concentrations is the presence of anions, such as phosphate (as P) and sulfate (as $SO_4$), through competition for adsorption sites. Concentrations of these anions are relatively low in the sampled groundwater (90th percentiles are 0.02 mg/L and 30 mg/L, respectively), suggesting that their presence probably does not greatly affect arsenic concentrations.
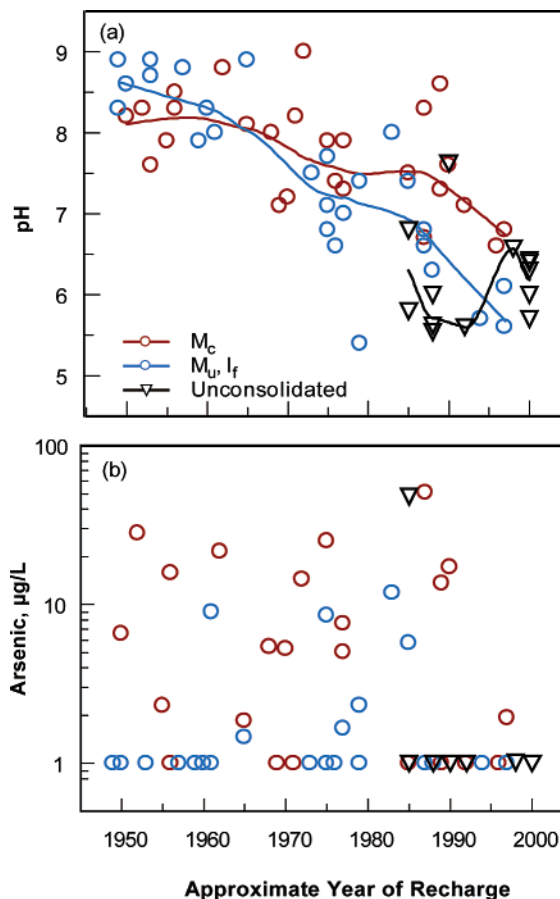
In samples from this study, high pH is related to the chemical evolution of the groundwater. Factors that affect the chemical evolution of the water include the age of the groundwater (residence time), the mineralogy of the aquifer materials, and the geochemistry of the groundwater system. The estimated date of recharge for water in the 58 bedrock wells sampled, based on a CFC concentration model (26), ranged from 1949 to 1997 (median is 1975), whereas the median date of recharge for the 30 unconsolidated aquifer samples was 1995 (Figure 5a). Generally, pH increases with increasing age of the sample (Figure 5a). Hydrolysis of silicate minerals, which are abundant in the bedrock and glacial materials of New England, is an important process that increases pH (17). Because these reactions do not rapidly reach equilibrium, the pH is generally greatest in older groundwater as suggested by groundwater ages estimated from the CFC content. This relation, however, varies by regional geology. For example, samples from the $M_c$ group have higher pH than similarly aged samples from the $M_u$ and $I_f$ group, except where samples are more than 40 years old. High pH in younger groundwater may reflect a greater abundance of calcite in the rocks of the $M_c$ group, although independent evidence for this has not been documented. High concentrations of arsenic are not directly related to the age of the water (Figure 5b), suggesting that other processes are related to increased pH.

Another process that may affect pH in bedrock aquifers in New England is cation exchange associated with past marine inundation. Whereas most groundwater with high arsenic concentrations is associated with rocks in the $M_c$ group, it is also within the area that was inundated by marine water during the Pleistocene glacial maximum (Figure 2). Water associated with rocks that were affected by marine
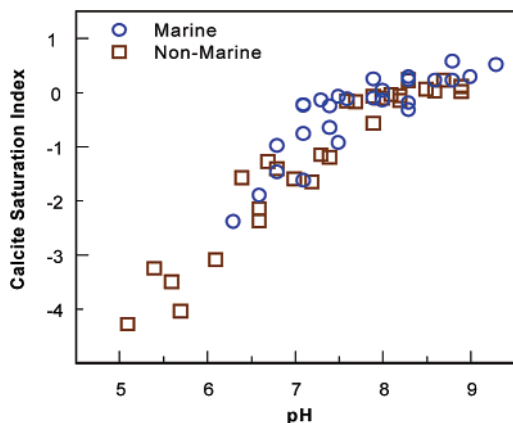
FIGURE 6. Relation between calcite saturation index and pH, inside and outside areas of Pleistocene marine inundation.
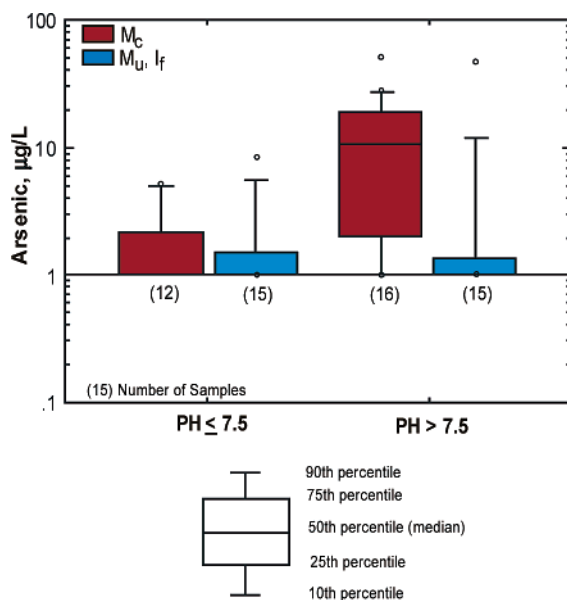


FIGURE 7. Distribution of arsenic concentrations by aquifer type and pH category.

TABLE 2. Estimated Population Supplied with Drinking Water with Arsenic Concentrations Greater than 10 $\mu$g/L in Eastern New England

| aquifer type (shown in Figure 2) | population, in thousands | percentage of wells with >10 $\mu$g/L of arsenic | estimated population supplied by wells with >10 $\mu$g/L of arsenic, in thousands |
|---|---|---|---|
| Public Drinking Water Wells | | | |
| unconsolidated aquifers | 2290 | 3 | 69 |
| bedrock aquifers | 199 | 9 | 18 |
| Private Drinking Water Wells | | | |
| bedrock aquifers ($M_c$) | 182 | 29 | 53 |
| bedrock aquifers ($M_u$, $I_f$) | 510 | 7 | 36 |
| bedrock aquifers (other) | 145 | 4 | 6 |
| unconsolidated aquifers | 250 | 3 | 8 |
| totals | 3576 | | 190 |

centrations are in areas with both high pH and specific geology ($M_c$).

The concentrations of arsenic in water in private bedrock wells also were evaluated for other water chemistry and physical variables. The occurrence and concentrations of arsenic is not strongly related to any other trace elements or to iron, manganese, specific conductance, sulfate, or sulfide. Data for well depth, overburden thickness, casing length, and yield were available for all wells, and, in general, arsenic concentrations is not directly related to any of these physical variables. Most of these chemical and physical variables are non-normally distributed, so Spearman's Rho, a nonparametric correlation coefficient, was computed (31). All correlation coefficients for arsenic and the chemical and physical variables were weak (less than 0.4) and none was significant at the $\alpha = 0.05$ confidence level. Local and regional anthropogenic factors (land use) may affect arsenic concentrations such as presence of landfills, which can alter the natural geochemistry of the underlying groundwater.

**Human Health and Water-Use Implications.** Public and private groundwater supplies in eastern New England can contain high levels of arsenic. All public water suppliers are required to meet the new arsenic standard of 10 $\mu$g/L by 2006 (3). The intent is to ensure that no one served by public water supplies will have arsenic concentrations greater than 10 $\mu$g/L. Private groundwater sources, in the region however, are not regulated and do not have to meet the Federal or State standards. Only citizens taking corrective actions will reduce the number of people on private groundwater sources with arsenic concentrations greater than 10 $\mu$g/L.

Data on concentrations of arsenic in groundwater in combination with information on water use in the region were used to estimate the number of people using groundwater for water supply that contains arsenic concentrations at levels of concern for human health. The estimates are intended only to represent the magnitude of the potential arsenic problem in the region; estimates of exposure are not presented and are beyond the scope of this study. The actual number of people with high-arsenic in their drinking water may be lower than estimated because of many factors, including (1) use of bottled water for drinking water and (2) water treatment systems that are effective in removing or reducing arsenic concentrations. The estimates were made on the basis of the percentages of wells with arsenic concentrations greater than 10 $\mu$g/L—the current USEPA standard—by aquifer type. The percentages are from results of this study and those reported in the associated retrospective study (7), along with water-use data from those same aquifer areas or types (Table 2).
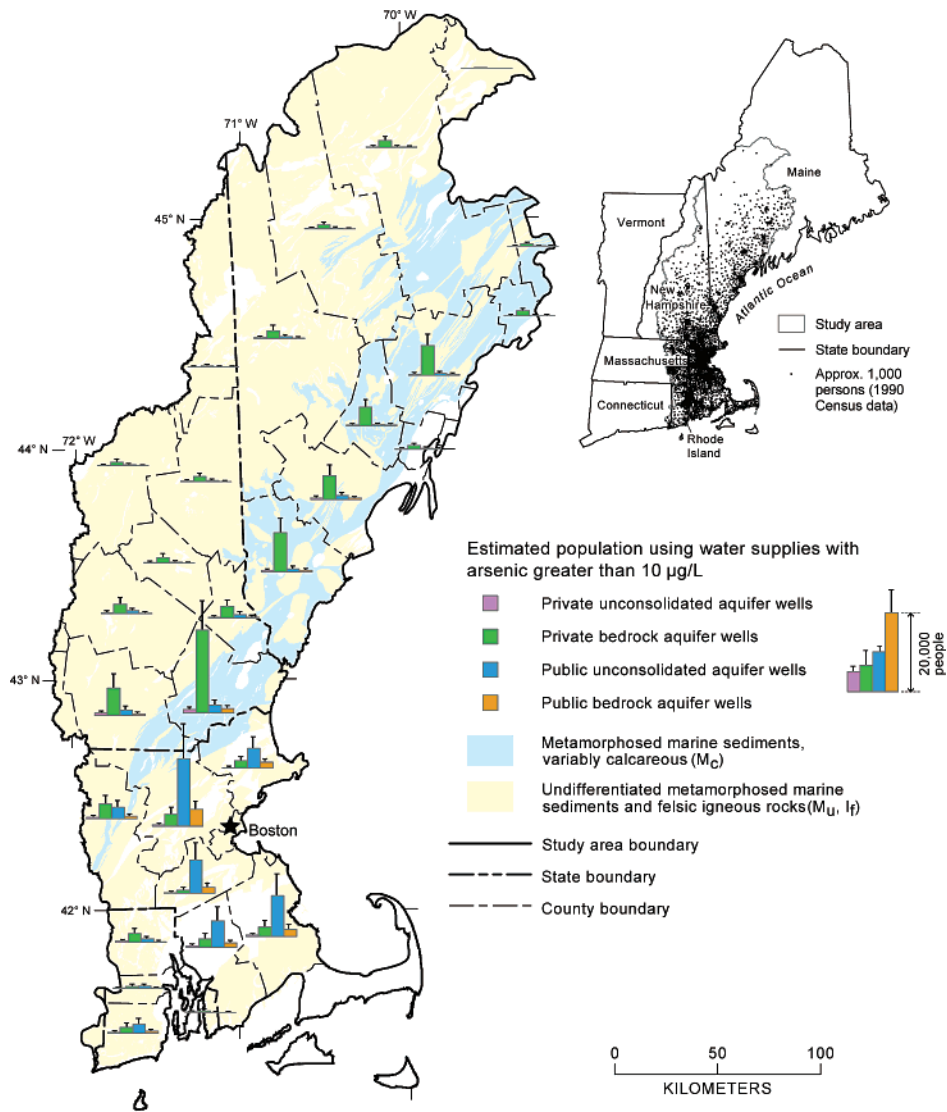
inundation tends to have high pH values and is more commonly in equilibrium with calcite compared to groundwater in aquifers unaffected by marine water (Figure 6). This greater tendency for a high pH (and calcite saturation) may be related to the exchange of calcium in groundwater for sodium on exchange sites in the aquifer solids. The process of cation exchange after flushing of seawater is well documented (41); by decreasing the aqueous calcium concentrations, the pH is raised by calcite dissolution. This process has been used to explain high pH in the Central Oklahoma aquifer where fresh groundwater displaced deep saline groundwater (42, 43). A recently suggested arsenic-release process, which may be relevant to eastern New England, is the carbonation of sulfide minerals (44); however, this process was not evaluated in the current study.

Additionally, groundwater in the unaffected aquifers (largely the $M_u$ and $I_f$ group) appears to be somewhat older (Figure 5a) compared to groundwater with an equivalent pH in marine-affected area of New England (largely the $M_c$ group). This might suggest that older groundwater from the $M_u$ and $I_f$ group would contain high arsenic concentrations; however, high arsenic does not always occur with high pH. This further implies the absence of an available arsenic source. For example, in Figure 7 the highest arsenic con-

**FIGURE 8.** Estimated number of people using water supplies with arsenic greater than 10 $\mu$g/L, by county and type of supply. Error bars represent asymptotic standard error.

The data on population using public and private groundwater supplies are derived from previous studies in the area and are shown in Table 2 (7, 21). For the water-use data, the following assumptions were made: (1) The percentage of public wells in unconsolidated aquifers with arsenic concentrations greater than 10 $\mu$g/L (3%) is a generalized rate and represents water delivered to homes for the entire study area; however, it may vary geographically with hydrologic and geologic conditions. (2) To estimate the population with private drinking-water wells by geologic group, the county-level water-use population data were assumed to be uniformly distributed across each county and, thus, could be apportioned into aquifer areas using GIS techniques. In addition, 77% of private drinking water wells are in bedrock (adjusted from 1990 census data) (20).

Water from private wells completed in bedrock aquifers is most likely to contain arsenic but supplies only about 30% of the groundwater use in the study area. Public supply wells in unconsolidated aquifers are much less likely to contain arsenic but supply about 70% of groundwater used for drinking water. The estimated number of people using water supplies with arsenic concentrations exceeding 10 $\mu$g/L is largest for those on private supplies—about 103 000 compared to 87 000 on public supplies (Table 2).

The private supply population most affected is in the eastern counties of Maine and New Hampshire and is related to private bedrock-well use (Figure 8). These counties are dominantly underlain by rocks of the $M_c$ group, which contain wells with the largest concentrations of arsenic in groundwater. Nearly 30% of the approximately 182 000 people in the $M_c$ aquifer area (about 53 000 people) using private bedrock wells may have water supplies with arsenic concentrations exceeding 10 $\mu$g/L (Table 2). In the western counties of Maine and New Hampshire as well as some counties in Massachusetts and Rhode Island private wells are in bedrock formations that are less likely to contribute arsenic to that well water, but collectively, these wells supply water to a large number of people.

This analysis of water use and arsenic concentration exceedence rates indicates that across eastern New England, more than 103 000 people with private wells may have water supplies with elevated arsenic concentrations. These private supplies, however, are not regulated or mandated to reduce arsenic concentrations and thus may continue to provide residents with water containing arsenic greater than the current drinking water standard unless action is taken by individual well owners. The geography of the high arsenic area also is of concern. The largest number of people with high arsenic concentrations in private supplies is in the eastern part of the region, where most of the population is located (Figure 8, inset map). Furthermore, because this part of the region is among the fastest growing part of New

England, private supplies continue to be developed in areas where arsenic concentrations frequently occur at levels greater than 10 $\mu$g/L.

The estimated number of people on public groundwater supplies with arsenic concentrations above 10 $\mu$g/L (87 000 people) is greatest in eastern Massachusetts (Table 2). However, this is a regulated resource, and by 2006, all public water suppliers will be required to provide water with arsenic below 10 $\mu$g/L.

## Acknowledgments

## Supporting Information Available

Table of data including major ions, trace elements, nutrients, chlorofluorocarbons, radionuclides, dissolved gases, field parameters, and ancillary data. This material is available free of charge via the Internet at http://pubs.acs.org.

## Literature Cited

(1) Focazio, M. J.; Welch, A. H.; Watkins, S. A.; Helsel, D. R.; Horn, M. A. *A Retrospective Analysis on the Occurrence of Arsenic in Ground Water Resources of the United States and limitations in Drinking-Water-Supply Characterizations*; U.S. Geological Survey Water-Resources Investigations Report 99-4279; 1999.

(2) Welch, A. H.; Westjohn, D. B.; Helsel, D. R.; Wanty, R. B. *Ground Water* **2000**, *38*, 589−604.

(3) U.S. Environmental Protection Agency, EPA to Implement 10 ppb Standard for Arsenic in Drinking Water. http://www.epa.gov/safewater/ars/ars-oct-factsheet.html (accessed Jan 2002).

(4) U.S. Environmental Protection Agency, National Primary Drinking Water Regulations; Arsenic and Clarifications to Compliance and New Source Contaminants Monitoring; Final Rule. http://www.epa.gov/safewater/ars/arsenic_finalrule.html (accessed Jan 2002).

(5) Loiselle, M. C.; Marvinney, R. G.; Smith, A. E. *Spatial Distribution of Arsenic in Groundwater in Maine*; Geological Society of America, 2001 Annual Meeting; Boston, MA, 2001; Geological Society of America: November 5−8, 2001.

(6) Karagas, M. R.; Le, C. X.; Morris, S.; Blum, J.; Lu, X.; Spate, V.; Carey, M.; Stannard, V.; Klaue, B.; Tosteson, T. D. *Int. J. Occup. Med. Environ. Health* **2001**, *14*, 171−175.

(7) Ayotte, J. D.; Nielsen, M. G.; Robinson, G. R., Jr.; 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. http://water.usgs.gov/pubs/wri/wri994162.

(8) Hon, R.; McTigue, D.; Stein, C.; Brandon, W. C. *Arsenic in Bedrock and Bedrock Groundwater in North Central Massachusetts*; Geological Society of America, Northeast Section, 36th Annual Meeting; Burlington, Vermont, 2000; Geological Society of America: March 12−14, 2001.

(9) Peters, S. C.; Blum, J. D.; Klaue, B.; Karagas, M. R. *Environ. Sci. Technol.* **1999**, *33*, 1328−1333.

(10) Reeve, A. S.; Horesh, M.; Warner, B.; Yates, M. *Geochemical evaluation of sources for arsenic in ground water, Northport, Maine*; Geological Society of America, Northeast Section, 36th Annual Meeting; Burlington, VT, 2000; Geological Society of America: March 12−14, 2001.

(11) Grantham, D. A.; Jones, J. F. *Am. Water Works Assoc. J.* **1977**, *69*, 653−657.

(12) Hinkle, S. R.; Polette, D. J. *Arsenic in Ground Water of the Willamette basin*; U.S. Geological Survey Water-Resources Investigations Report 98-4025; 1999.

(13) Warner, K. L. *Ground Water* **2001**, *39*, 433−442.

(14) Chormann, F. H., Jr., Masters Thesis, Department of Earth Sciences, University of New Hampshire, Durham, 1985.

(15) D'Angelo, D.; Norton, S. A.; Loiselle, M. C. *Historical Uses and Fate of Arsenic in Maine*; Water Research Institute Completion Report; 1996.

(16) Hem, J. D. *Study and Interpretation of the Chemical Characteristics of Natural Water*; U.S. Geological Survey Water-Supply Paper 2254; 1985.

(17) Rogers, R. J. *Ground Water* **1989**, *27*, 690−712.

(18) Robinson, G. R., Jr. *Portraying Chemical Properties of Bedrock for Water Quality and Ecosystem Analysis: an Approach for the New England Region*; U.S. Geological Survey Open-File Report 97-154; 1997.

(19) U.S. Geological Survey, Aggregate Water-Use Data System (AWUDS). http://water.usgs.gov/watuse/wuawuds.html (accessed Oct 2001).

(20) U.S. Census Bureau, Historical Census of Housing Tables − Sources of Water. http://www.census.gov/hhes/www/housing/census/historic/water.html (accessed Oct 2002).

(21) Flanagan, S. M.; Nielsen, M. G.; Robinson, K. W.; Coles, J. F. *Water Quality Assessment of the New England Coastal Basins in Maine, Massachusetts, New Hampshire, and Rhode Island-Environmental Settings and Implications for Water Quality and Aquatic Biota*; U.S. Geological Survey Water-Resources Investigations Report 98-4249; 1999.

(22) Robinson, G. R., Jr.; Ayotte, J. D.; Montgomery, D. L.; DeSimone, L. A. *Lithogeochemical Character of Near-Surface Bedrock in the New England Coastal Basins*; U.S. Geological Survey Open-File Report 02-7; 2002.

(23) Montgomery, D. L.; Robinson, G. R., Jr.; Robinson, K. W.; Ayotte, J. D.; Flanagan, S. M. *Digital Data set of Generalized Lithogeochemical Characteristics of Near-Surface Bedrock in the New England Coastal Basins*; U.S. Geological Survey Water Fact Sheet FS-003-02; 2002.

(24) Scott, J. C. *Computerized Stratified Random Site-Selection Approaches for Design of Ground Water Quality Sampling Networks*; U.S. Geological Survey Water-Resources Investigations Report 90-4101; 1990.

(25) Ayotte, J. D.; Ryker, S. J.; Proctor, A. L. *Variation in Bedrock Ground Water Arsenic Concentrations and Implications for Human-health Sudies in New England*; Geological Society of America, 2001 Annual Meeting; Boston, MA, 2001; Geological Society of America: November 5−8, 2001.

(26) Busenberg, E.; Plummer, L. N. *Concentrations of Chlorofluorocarbons and other Gases in Groundwaters at Mirror Lake, New Hampshire*; U.S. Geological Survey Toxics Substances Hydrology Program-Proceedings of the Technical Meeting; Colorado Springs, CO, 1993; U.S. Geological Survey Water-Resources Investigations Report 94-4015; September 20−24, 1993.

(27) Koterba, M. T.; Wilde, F. D.; Lapham, W. W. *Ground Water Data-Collection Protocols and Procedures for the National Water-Quality Assessment Program- -Collection and Documentation of Water-Quality Samples and Related Data*; U.S. Geological Survey Open-File Report 95-399; 1995.

(28) Lapham, W. W.; Wilde, F. D.; Koterba, M. T. *Ground Water Data-Collection Protocols and Procedures for the National Water-Quality Assessment Program- -Selection, Installation, and Documentation of Wells, and Collection of Related Data*; U.S. Geological Survey Open-File Report 95-398; 1995.

(29) Garbarino, J. R. *Methods of Analysis by the U.S. Geological Survey National Water Quality Laboratory- -Determination of Whole-Water Recoverable Arsenic, Boron, and Vanadium using Inductively Coupled Pasma-Mass Spectrometry*; U.S. Geological Survey Open-File Report 99-464; 2000.

(30) Garbarino, J. R. *Methods of Analysis by the U.S. Geological Survey National Water Quality Laboratory- -Determination of Dissolved Arsenic, Boron, Lithium, Selenium, Strontium, Thallium, and Vanadium using Inductively Coupled Plasma-Mass Spectrometry*; U.S. Geological Survey Open-File Report 99-093; 1999.

(31) Helsel, D. R.; Hirsch, R. M. *Statistical Methods in Water Resources*; Elsevier Science Company, Inc.: New York, 1992.

(32) Osberg, P. H.; Hussey, A. M. I.; Boone, G. M. *Bedrock Geologic Map of Maine*; Maine Geological Survey: 1985.

(33) Robinson, G. R., Jr., written communication.

(34) Ayuso, R. A.; Foley, N. K.; Ayotte, J. D.; Lyon, A.; Burns, J.; Marvinney, R. G.; Reeve, A. S.; Robinson, G. R., Jr. *Lead Isotopes, Arsenic Sources and Enrichment Pathways Linking Sulfides from Mines and Unmineralized Rocks to Secondary Iron Oxides, Coastal New England*; Arsenic in New England: A Multidisciplinary Scientific Conference; Manchester, NH, 2002; New Hampshire Consortium on Arsenic: May 29−31, 2002.

(35) Ayuso, R. A., written communication.

(36) Bhumbla, D. K.; Keefer, R. F. In *Arsenic in the Environment, Part 1: Cycling and Characterization*; Nriagu, J. O., Ed.; John Wiley & Sons: New York, 1994; Vol. 26, p 430.

(37) Smedley, P. L.; Kinniburgh, D. G. *Appl. Geochem.* **2002**, *17*, 517−568.

(38) Smedley, P. L.; Kinniburgh, D. J.; Huq, I.; Zhen-dong, L.; Nicolli, H. B. In *Arsenic Exposure and Health Effects IV*, 1st ed.; Chappell, W. R., Abernathy, C. O., Calderon, R. L., Eds.; Elsevier Science Ltd.: New York, 2001; p 467.

(39) Stollenwerk, K. G. In *Arsenic in Ground Water: Geochemistry and Occurrence*; Welch, A. H., Stollenwerk, K. G., Eds.; Kluwer Academic Publishers: Boston, 2003; pp 67−100.

(40) Dzombak, D.; Morel, F. *Surface Complexation Modeling: Hydrous Ferric Oxide*; Wiley: New York, 1990.

(41) Appelo, C. A. J.; Postma, D. *Geochemistry, Groundwater and Pollution*; A. A. Balkema: Rotterdam, 1993.

(42) Parkhurst, D. L.; Appelo, C. J. *Users guide to PHREEQC (version 2): A Computer Program for Speciation, Batch Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations*; U. S. Geological Survey Water-Resources Investigations Report 99-4259; 1999.

(43) Schlottmann, J. L.; Breit, G. N. In *Water-Rock Interaction*; Kharaka, Y. K., Maest, A. S., Eds.; A. A. Balkema: Rotterdam, 1992; pp 835−838.

(44) Kim, M. J.; Nriagu, J.; Haack, S. *Environ. Sci. Technol.* **2000**, *34*, 3094−3100.