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Worldwide Occurrences of Arsenic in Ground Water

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As the world population increases beyond 6 billion, one of the most fundamental resources for human survival, clean water, is decreasing. Revised estimates from the World Health Organization for 1990 indicate that 43% of the world's population do not have adequate sanitation and 22% do not have clean drinking water (*1, 2*). The rising demands for sanitary water often cannot be met by surface-water supplies. This has led to increased dependence on ground-water resources in many parts of the world. The consequences of ground-water development often include overdrafting, land subsidence, and the use of ground water unfit for human consumption. The recent increased utilization of ground water in India and Bangladesh has caused new health issues (*3–5*). An estimated 36 million people in the Bengal Delta are at risk from drinking arsenic-contaminated water. Numerous other occurrences worldwide have been reported (see table, right) (*6*), and some of these, such as those in Taiwan, have been recognized for several decades.

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Arsenic is not found in high abundance in the Earth's continental crust; it is less abundant than several of the "rare-earth" elements (*7*). Unlike the rare-earth elements, however, arsenic is commonly concentrated in sulfide-bearing mineral deposits, especially those associated with gold mineralization, and it has a strong affinity for pyrite (*8*), one of the more ubiquitous minerals in the Earth's crust. It is also concentrated in hydrous iron oxides. Arsenic can be easily solubilized in ground waters depending on pH, redox conditions, temperature, and solution composition (*6*). Many geothermal waters contain high concentrations of arsenic (*9*). Natural arsenic in ground water at concentrations above the drinking water standard of 10 μg/liter is not uncommon. Man-made sources of arsenic, such as mineral extraction and processing wastes, poultry and swine feed additives, pesticides, and highly soluble arsenic trioxide stockpiles are also not uncommon and have caused the contamination of soils and ground waters.

A small number of source materials are now recognized as significant contributors to arsenic in water supplies: organic-rich or black shales, Holocene alluvial sediments with slow flushing rates, mineralized and mined areas (most often gold deposits), volcanogenic sources, and thermal springs. The relationship between high arsenic concentrations and geothermal waters is not a simple one. Arsenic concentrations are high in the thermal waters of Kamchatka, New Zealand, Japan, Alaska, California, and Wyoming, where black shales are common, but they are low in thermal waters from Hawaii and Iceland (*10*), where most of the rocks are geologically young basalts. Aquifers with carbonaceous shales and without obvious thermal gradients, such as in Taiwan, also can lead to high dissolved arsenic concentrations.

Two other environments can lead to high arsenic: (i) closed basins in arid-to-semi-arid climates (especially in volcanogenic provinces) and (ii) strongly reducing aquifers, often composed of alluvial sediments but with low sulfate concentrations. Young sediments in low-lying regions of low hydraulic gradient are characteristic of many arsenic-rich aquifers. Ordinary sediments containing 1 to 20 mg/kg (near crustal abundance) of arsenic can give rise to high dissolved arsenic (>50 μg/liter) if initiated by one or both of two possible "triggers"—an increase in pH above 8.5 or the onset of reductive iron dissolution (*6*). Potentially important, additional factors promoting arsenic solubility are high concentrations of phosphate, bicarbonate, silicate, and/or organic matter in the ground waters. These solutes can decrease or prevent the adsorption of arsenate and arsenite ions onto fine-grained clays, especially iron oxides. Arsenite tends to adsorb less strongly than arsenate often causing arsenite to be present at higher concentrations. Unfortunately, these generalities do not allow prediction of high or low dissolved arsenic concentrations in any particular well because of heterogeneous distributions in the aquifers. Furthermore, arsenic concentrations can change in any given well over the course of a few years so that regular monitoring is required in high-risk areas (*11*).

GLOBAL ARSENIC CONTAMINATION IN GROUND WATER

Country/ region	Potential exposed population	Concentration (μg/liter)	Environmental conditions
Bangladesh	30,000,000	<1 to 2,500	Natural; alluvial/deltaic sediments with high phosphate,* organics
West Bengal, India	6,000,000	<10 to 3,200	Similar to Bangladesh
Vietnam	>1,000,000	1 to 3,050	Natural; alluvial sediments
Thailand	15,000	1 to >5,000	Anthropogenic; mining and dredged alluvium
Taiwan[†]	100,000 to 200,000	10 to 1,820	Natural; coastal zones, black shales
Inner Mongolia	100,000 to 600,000	<1 to 2,400	Natural; alluvial and lake sediments; high alkalinity
Xinjiang, Shanxi	>500	40 to 750	Natural; alluvial sediments
Argentina	2,000,000	<1 to 9,900	Natural; loess and volcanic rocks, thermal springs; high alkalinity
Chile[‡]	400,000	100 to 1,000	Natural and anthropogenic; volcanogenic sediments; closed basin lakes, thermal springs, mining
Bolivia[§]	50,000	–	Natural; similar to Chile and parts of Argentina
Brazil[‖]	–	0.4 to 350	Gold mining
Mexico	400,000	8 to 620	Natural and anthropogenic; volcanic sediments, mining
Germany	–	<10 to 150	Natural: mineralized sandstone
Hungary, Romania	400,000	<2 to 176	Natural; alluvial sediments; organics
Spain[¶]	>50,000	<1 to 100	Natural; alluvial sediments
Greece[#]	150,000	–	Natural and anthropogenic; thermal springs and mining
United Kingdom[**]	–	<1 to 80	Mining; southwest England
Ghana	<100,000	<1 to 175	Anthropogenic and natural; gold mining
USA and Canada	–	<1 to >100,000	Natural and anthropogenic; mining, pesticides, As_2O_3 stockpiles, thermal springs, alluvial, closed basin lakes, various rocks

[*] These estimates [from (*6*) except where noted] are highly uncertain, difficult to obtain, and changing as new water sources or treatment are established. [†] Additional estimate from (*15, 16*). [‡] Installation of a new treatment plant has greatly decreased the exposed population. [§] Estimate from (*16*). [‖] Source (*17*), no ground waters analyzed. [¶] Source (*18*). [#] Source (*6*) and (*16*). [**] Source (*19*)

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In a number of areas worldwide, oxidation and dissolution of arsenian pyrite, $Fe(As,S)_2$, and arsenopyrite, FeAsS, are additional processes that lead to high concentrations of dissolved arsenic (*12*). The oxidation can be promoted naturally through infiltrating oxygenated ground waters (*13*) or through lowering of the ground-water table (by well-water pumping or climate variations) into a stratigraphic zone containing arsenic-rich sulfides (*14*). The highest natural arsenic concentrations found in the United States (1 to 10 mg/liter) are in the Fairbanks, Alaska, area, where arsenopyrite-rich zones in igneous and metamorphic rocks are being oxidized, and there may also be some iron reduction (*13*).

The key to minimizing risk is to incorporate hydrogeological, geochemical, and microbiological expertise into the decision-making process of water managers, remediation specialists, and policy-makers. The geologic and ground-water conditions that promote high arsenic concentrations are known and can help identify high-risk areas. The western United States has many ground waters where arsenic is found in concentrations >10 μg/liter, and treating them will be expensive but may be trivial compared with potential health-care costs. In the search for adequate water supplies and in the absence of adequate information, it is prudent to test selected wells before opening the tap.

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Arsenic Epidemiology and Drinking Water Standards

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In the United States, setting the maximum contaminant level (MCL) that regulates the concentration of arsenic in public water supplies has been an extraordinarily protracted process (see the table on this page). Recently, the MCL was lowered to 10 μg/liter, from the 50 μg/liter standard established in 1942. However, as early as 1962 the USPHS advised that water concentrations should not exceed 10 μg/liter when "more suitable supplies are or can be made available" (*1*). In 1986, Congress directed the U.S. Environmental Protection Agency (EPA) to revise the standard by 1989, but it failed to do so (*2*). Not until January 2001, in one of the last acts of the Clinton administration, was the announcement of a new U.S. standard of 10 μg/liter made by the EPA (*3*). Two months later, the Bush administration delayed adoption of the standard, citing concerns about the science supporting the rule and its estimated cost (*2*). Nevertheless, in October 2001, under pressure from Congress and following a pivotal report by the National Research Council (NRC) (*4*), the EPA adopted the 10 μg/liter standard (*2*) (see the table, below). We will consider how the regulatory process might interpret and respond more effectively to results from epidemiological studies.

Arsenic was one of the first chemicals recognized as a cause of cancer. As early as 1879, the high rates of lung cancer in miners in Saxony were attributed in part to inhaled arsenic (*5*). A few years later, skin cancers were reported in patients treated with medicine containing arsenic (*6, 7*). Evidence that arsenic in drinking water could cause skin cancer came much later, in the 1930s, from Argentina (*8*), and subsequently from many other countries (*9*), including a large population in Taiwan (*10*).

In the 1960s, evidence emerged in Argentina that arsenic in drinking water might cause internal cancers, particularly of the lung and urinary tract (*11, 12*). Startling results from Taiwan, appearing in 1985, showed increased mortality from several cancers, especially lung, bladder, and kidney cancers (*13*). Bladder cancer mortality rates for those with more than 600 μg/liter of arsenic in their water were more than 30 to 60 times the rates in the unexposed population (*14*). Such high cancer rates were unprecedented for any water contaminant. By 1992, the combination of evidence from Taiwan and elsewhere was

HISTORY OF U.S. STANDARDS FOR ARSENIC IN DRINKING WATER

1942	USPHS sets an interim drinking water standard of 50 μg As/liter (*50*)
1962	USPHS identifies 10 μg As/liter as the goal (*1*)
1975	EPA adopts the interim standard of 50 μg As/liter set by the USPHS in 1942 (*50*)
1986	Congress directs EPA to revise the standard by 1989 (*2*)
1988	EPA estimates that the ingestion of 50 μg As/liter results in a skin cancer risk of 1 in 400 (*51*)
1992	Internal cancer risk estimated to be 1.3 per 100 persons at 50 μg As/liter (*16*)
1993	World Health Organization recommends lowering arsenic in drinking water to 10 μg As/liter (*52*)
1996	Congress directs the EPA to propose a new drinking water standard by January 2000 (*2*)
1999	NRC estimates cancer mortality risks to be about 1 in 100 at 50 μg As/liter (*28*)
2000	EPA proposes a standard of 5 μg As/liter and requests comment on 3, 10, and 20 μg As/liter (*2*)
2001	(January) Clinton EPA lowers the standard to 10 μg As/liter (*2*)
2001	(March) Bush EPA delays lowering the standard (*2*)
2001	(September) New NRC report concludes that EPA underestimated cancer risks (*4*)
2001	(October) EPA announces it will adopt the standard of 10 μg/liter (*2*)
2002	(February) The effective date for new standard of 10 μg As/liter (*2*)
2006	Compliance date for the new arsenic standard (*2*)

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