Source, Distribution, and Management of Arsenic in Water from Wells, Eastern San Joaquin Ground-Water Subbasin, California

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Summary

Between 1974 and 2001 water from as many as one-third of wells in the Eastern San Joaquin Ground-Water Subbasin, about 80 miles east of San Francisco, had arsenic concentrations greater than the U.S. Environmental Protection Agency Maximum Contaminant Level (MCL) for arsenic of 10 micrograms per liter (µg/L). Water from some wells had arsenic concentrations greater than 60 µg/L. The sources of arsenic in the study area include (1) weathering of arsenic bearing minerals, (2) desorption of arsenic associated with iron and manganese oxide coatings on the surfaces of mineral grains at pH's greater than 7.6, and (3) release of arsenic through reductive dissolution of iron and manganese oxide coatings in the absence of oxygen. Reductive dissolution is responsible for arsenic concentrations greater than the MCL. The distribution of arsenic varied areally and with depth. Concentrations were lower near ground-water recharge areas along the foothills of the Sierra Nevada; whereas, concentrations were higher in deeper wells at the downgradient end of long flow paths near the margin of the San Joaquin Delta (fig. 1). Management opportunities to control high-arsenic

concentrations are present because water from the surface discharge of wells is a mixture of water from the different depths penetrated by wells. On the basis of well-bore flow and depth-dependent water-quality data collected as part of this study, the screened interval of a public-supply well having arsenic concentrations that occasionally exceed the MCL was modified to reduce arsenic concentrations in the surface discharge of the well. Arsenic concentrations from the modified well were about 7 µg/L. Simulations of ground-water flow to the well showed that although upward movement of high-arsenic water from depth within the aquifer occurred, arsenic concentrations from the well are expected to remain below the MCL.
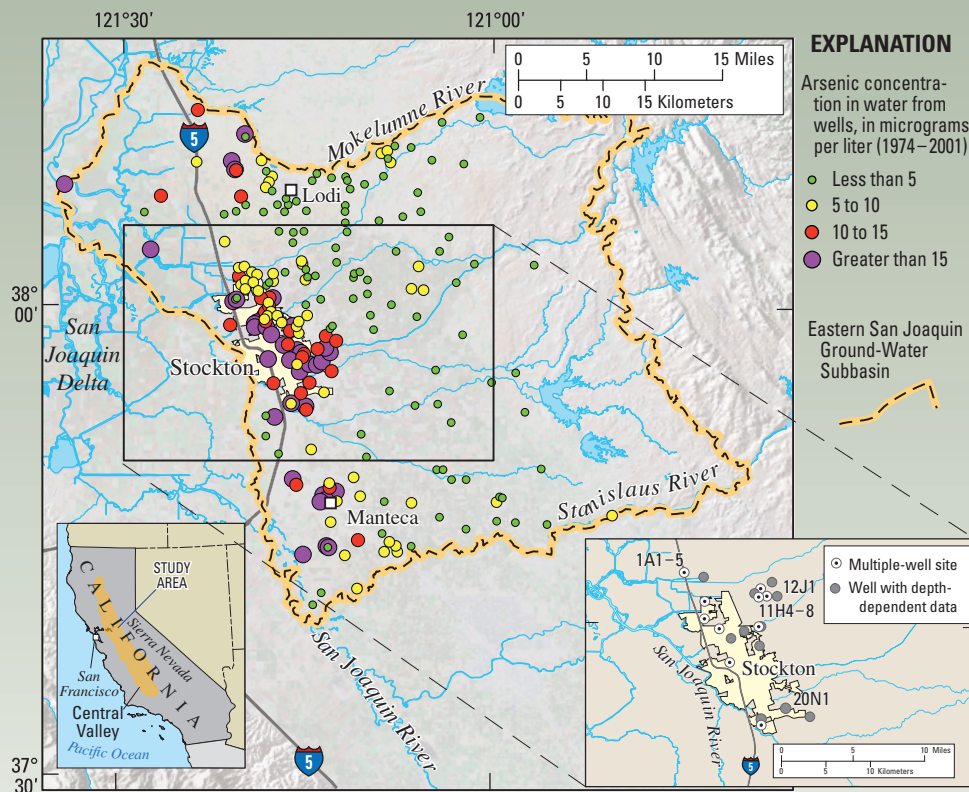
Figure 1. Arsenic concentrations in water from wells (1974-2001), multiple-well sites, and wells having depth-dependent data (2005-2008), Eastern San Joaquin Ground-Water Subbasin, California.

Introduction

Arsenic occurs naturally in the earth's crust and concentrations in water can be as high as 5,000 µg/L (Smedley and Kinniburgh, 2002). High concentrations in ground water may be present as a result of (1) geologic sources such as the weathering of sulfide minerals, (2) geochemical conditions in oxic (oxygen present) alkaline ground water, and in reducing (oxygen absent) ground water, (3) evaporation in closed basins in arid areas, or (4) in association with volcanic rocks or geothermal waters (Welch and Stollenwerk, 2003). Arsenic can be present in water as either arsenite (As III) or arsenate (As V). As(III) is more toxic than As(V)—although both forms are toxic, and the MCL for drinking water is for total arsenic.

In January 2006, the U.S. Environmental Protection Agency MCL for arsenic in drinking water was reduced from 50 to 10 micrograms per liter (µg/L) in response to recent information concerning the risk of lung and bladder cancers (Morales and others, 2000). About 5 percent of water systems in the United States, serving more than 11 million people, were expected to exceed the new MCL (Tienmann, 2001). Arsenic in excess of the MCL is of particular concern for ground-water supplies in parts of New England, the interior plains, and the western United States (Federal Register, 2001; Welch and others, 2000). Large spatial variability in arsenic concentrations within aquifers often makes it difficult to predict where high-arsenic water will be encountered (Smedley and Kinniburgh, 2002). Increased knowledge of the occurrence and distribution of arsenic in ground-water, especially its distribution with depth, may lead to management strategies and changes in well-construction practices designed to avoid the pumping of high-arsenic ground water. This may reduce the need for arsenic removal by expensive treatment, or the need to blend water from different sources to reduce arsenic concentrations in water served to consumers. Modification of existing wells to exclude high-arsenic ground-water may reduce

the need to abandon wells that do not meet the current MCL.

The purpose of this paper is to present preliminary (2008) results from an ongoing study of geochemistry and recharge to wells in the Eastern San Joaquin Ground-Water Subbasin. The scope of the study included test-drilling, geophysical logging, water-quality data collection, well-bore flow logging, and depth-dependent water-quality sampling. The study approach coupled a basin-wide areal assessment of arsenic concentrations with more detailed data collected from long-screened wells in areas with high-arsenic concentrations. The study evaluated the modification of a well to exclude water from depths having undesirable concentrations of arsenic as a management option to reduce arsenic concentrations in water from public-supply wells. A two-dimensional, radial ground-water flow model was developed to evaluate the long-term effectiveness of the well modification.

Hydrogeology

The study area is the Eastern San Joaquin Ground-Water Subbasin near Stockton, California, about 80 miles (mi) east of San Francisco (fig. 1). The ground-water subbasin is about 1,100 mi^2 (California Department of Water Resources, 2003), and is part of the larger San Joaquin Ground-Water Basin that forms the southern two-thirds of the Central Valley of California. The climate is characterized by hot, dry summers, and cool, moist winters. Average annual precipitation ranges from about 10 to 18 inches. Precipitation is greater in the Sierra Nevada, to the east of the study area. Runoff from these mountains, primarily as snowmelt, sustains flows in the Mokelumne and Stanislaus Rivers that bound the study area to the north and south. The San Joaquin River, which drains the San Joaquin Valley from south to north, bounds the study area to the west, and the foothills of the Sierra Nevada bound the study area to the east (fig. 1).

The study area is underlain by several thousand feet of consolidated,

partly-consolidated, and unconsolidated sedimentary deposits (California Department of Water Resources, 1967). The oldest deposits are marine and generally contain saline water, except where they crop out near the foothills of the Sierra Nevada. Regional ground-water flow in the San Joaquin Valley is through these marine deposits from recharge areas in the south to discharge areas near the San Joaquin delta. The marine deposits are separated from overlying sedimentary deposits by low-permeability volcanic deposits known as the Mehrten formation (Curtis, 1954). The volcanic deposits are about 1,000 feet (ft) below land surface in the Stockton area, and at shallower depths to the east toward the foothills of the Sierra Nevada. In the study area, the volcanic deposits are blanketed by a layer of alluvium eroded from the volcanic deposits (California Department of Water Resources, 1967). The overlying sedimentary deposits are divided into alluvial deposits eroded from the Sierra Nevada and its foothills, and the fluvial and delta deposits of the San Joaquin River.

Under predevelopment conditions, the direction of ground-water flow in the alluvial deposits was from recharge areas in the east toward discharge areas near the San Joaquin delta. Within the study area, ground-water recharge to these deposits is about 900,000 acre-feet per year (acre-ft/yr) (CDM, Inc., 2001). Pumping for municipal and agricultural supply exceeds recharge by 150,000 acre-ft/yr (CDM, Inc., 2001), and a large regional pumping depression has developed east of the city of Stockton. In 2000, the study area had a population of about 580,000 and population is expected to increase to more than 1.2 million by 2040 (CDM, Inc., 2001).

Sources of Arsenic

In the study area, arsenic is weathered from minerals in volcanic deposits, in alluvium eroded from granitic rocks in the Sierra Nevada, and in alluvium eroded from metamorphic and volcanic rocks in the foothills of the Sierra Nevada. Sulfide minerals, associated
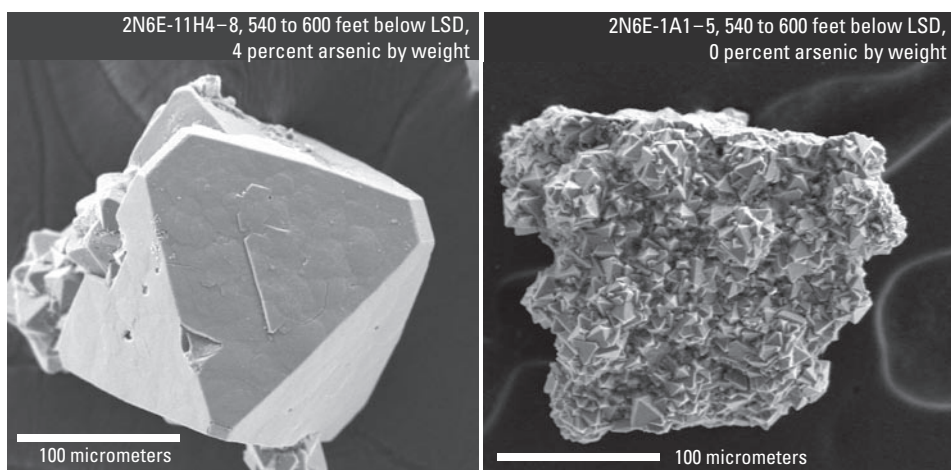
2N6E-11H4–8, 540 to 600 feet below LSD, 4 percent arsenic by weight

100 micrometers

2N6E-1A1–5, 540 to 600 feet below LSD, 0 percent arsenic by weight

100 micrometers

Figure 2. Scanning electron photomicrographs of pyrite crystals from selected drilling samples, Eastern San Joaquin Ground-Water Subbasin, California.

with metamorphic rocks and volcanic deposits in the foothills, are more abundant and contain greater amounts of arsenic than minerals in granitic rocks that compose the Sierra Nevada (Brown and Allan, 1955). As a consequence, smaller streams that drain the lower altitudes of the foothills may have eroded and deposited alluvium having greater abundance of arsenic than larger streams, such as the Mokelumne and Stanislaus Rivers, which drain the higher altitudes of the Sierra Nevada.

Iron-sulfide minerals, such as pyrite, commonly contain arsenic substituted within the crystal structure. Pyrite may originate as a primary mineral eroded from source rock or as a secondary mineral formed within aquifer deposits. Octahedral pyrite crystals (fig. 2), interpreted to be a primary mineral, were found in drilling samples collected from the volcanic deposits and alluvium eroded from those deposits. These crystals contained between 1 and 4 percent arsenic by weight. Some crystals show evidence of weathering and dissolution that may release arsenic to ground water. Twinned-pyritohedron crystals, interpreted to be a secondary mineral formed within aquifer deposits, also were found in drilling samples from the study area. Although incorporation of arsenic into secondary minerals could remove arsenic from ground water, arsenic was not detected within the crystal structure of secondary pyrite minerals (fig. 2).

Although the ultimate geologic source of arsenic is mineral weathering, arsenic sorbed to iron and manganese hydroxides on the surfaces of mineral grains may be more readily released to ground water than arsenic bound within the crystal structure of minerals. For example, under oxic conditions at alkaline pH's, arsenic may be mobilized into ground water by exchange with arsenic sorbed to iron and manganese hydroxide coatings on mineral surfaces (Welch and others, 2000). Under reducing conditions, arsenic may be mobilized into ground water as these coatings dissolve (Welch and others, 2000).

Acid-extractable arsenic concentrations provide a measure of the arsenic sorbed on mineral grains. Acid-extractable concentrations in core material and drill cuttings collected as part of this study ranged from 0.05 to 1.6 mg / kg of alluvium. These concentrations are not unusual compared to the average continental abundance of arsenic (Hem, 1985). Arsenic sorbed on the surface of mineral grains is a large reservoir of arsenic in the study area, with the highest concentrations in the fine-grained, organic-rich delta deposits. As much as 90 percent of the acid-extractable arsenic was in the oxidized form, with a median of 64 percent as As(V). Most of the reduced arsenic, As(III), was associated with either organic or sulfur complexes. Concentrations in ground water are not limited by the availability of arsenic

sorbed on mineral grains; instead specific geochemical conditions, especially redox and pH, within aquifers control arsenic concentrations in ground water.

Arsenic in Water from Wells

Occurrence and distribution of arsenic in public-supply and irrigation wells in the San Joaquin Valley of California was described by Belitz and others (2003). Although arsenic concentrations in most wells were generally less than 5 µg/L, about 13 percent of wells in the San Joaquin Valley had concentrations in excess of 10 µg/L. Belitz and others (2003) identified ground water near the margin of the San Joaquin Delta as having high-arsenic concentrations.

Arsenic concentrations in water from wells the Eastern San Joaquin Ground-Water Subbasin ranged from less than the reporting limit (1 to 4 µg/L, depending on the method of analysis) to 60 µg/L, with a median concentration of 6 µg/L (fig. 1). These data are based on 1,190 samples collected from more than 250 wells between 1974 and 2004 by federal, state, and local agencies. Only 2 samples were greater than the previous MCL for arsenic of 50 µg/L, while about 26 percent of the samples were greater than the MCL of 10 µg/L. Arsenic concentrations in samples collected from 50 wells as part of this study between May 2004 and February 2007 were similar and ranged from 1 to 63 µg/L with a median concentration of 5.1 µg/L. About one-third of those samples were greater than the MCL of 10 µg/L. Water from wells having arsenic concentrations greater than 10 µg/L can no longer be used as a source of public supply without treatment or blending.

Arsenic concentrations in water from wells were not uniformly distributed areally or with depth in the study area (fig. 1). Arsenic concentrations were less than 10 µg/L in the eastern part of the study area near ground-water recharge areas along the foothills of the Sierra Nevada. Arsenic concentrations increased to the west, and high concentrations were distributed in a northwest trending band near the margin of the San Joaquin Delta (fig. 1). This is similar to the distribution
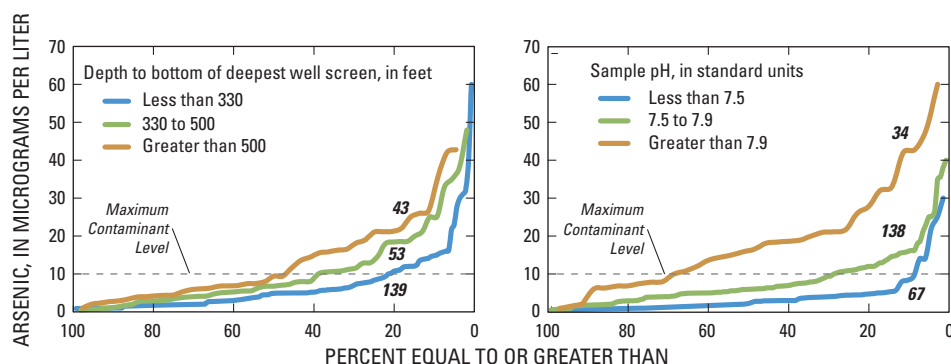
Figure 3. Cumulative distribution of average arsenic concentrations in water from wells as a function of well depth and pH, Eastern San Joaquin Ground-Water Subbasin, California, 1974 to 2004. Number in figure is numbers of wells within each group.

of high-chloride water in the study area (Izbicki and others, 2006).

Arsenic concentrations in water from the surface discharge of wells generally increased with well depth. The probability that water from a well would have an arsenic concentration greater than or equal to the MCL of 10 µg/L was 21 percent for wells less than 330 ft deep, 39 percent for wells between 330 and 500 ft deep, and 48 percent for wells greater than 500 ft deep (fig. 3). This relation with well depth varied spatially. For example, there was no correlation between well depth and arsenic concentrations in the eastern part of the study area where arsenic concentrations were generally low. In contrast, along the western side of the study area adjacent to the San Joaquin Delta, arsenic concentrations were commonly greater than 10 µg/L, regardless of well depth.

Well-bore flow and depth-dependent water-quality data were collected from 11 wells in the subbasin (fig. 1). Data from well 1N/7E-20N1 (fig. 4) illustrate the depth-dependence of arsenic concentrations in wells within the study area. Within well 20N1, about 55 percent of the water was yielded from the upper part of the screened interval between 158 and 204 ft below land surface. Water from these shallower depths had an arsenic concentration about 5 µg/L. In contrast, water from deeper depths had arsenic concentrations in excess of 15 µg/L (fig. 4). The higher concentrations were masked by mixing within the well as water was pumped to the surface. Water from well 20N1 has a history of arsenic

concentrations greater than the MCL of 10 µg/L, although at the time of sample collection well 20N1 had an arsenic concentration of 8.9 µg/L, slightly below the MCL. In contrast to arsenic, chloride and nitrate concentrations were higher in the upper part of well 20N1 (fig. 4). High-chloride and high-nitrate concentrations are probably the result of agricultural

activities that altered the quality of shallow ground water.

pH, Redox, and Microbiological Controls

Arsenic concentrations in water from the surface discharge of wells increased with pH (fig. 3). The probability that water from a well would have an arsenic concentration greater than or equal to the MCL of 10 µg/L was 7 percent if the pH was less than 7.5, 29 percent if the pH was between 7.5 and 7.9, and 69 percent if the pH was greater than 7.9. The pH dependence of arsenic is consistent with results of previous work in the San Joaquin Valley (Belitz and others, 2003). pH was not uniformly distributed throughout the study area, and was generally lower in the eastern part of the study area and higher in deep wells farther downgradient.
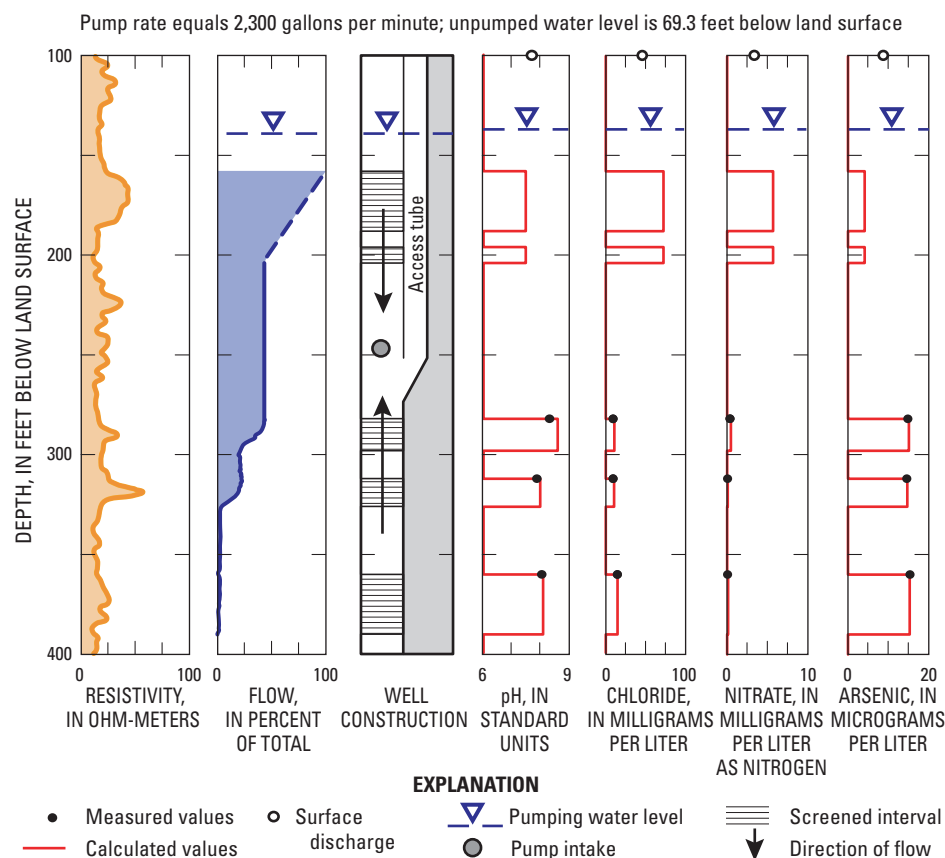

Figure 4. Well-bore flow and depth-dependent water-quality data from well 1N/7E-20N1, Eastern San Joaquin Ground-Water Subbasin, California, August 2004.

Arsenic concentrations in oxic water from wells were less than the MCL of 10 µg/L, and concentrations in these samples increased with pH (fig. 5), consistent with exchange of arsenic sorbed to iron and manganese hydroxides on mineral grains (Smedley and Kinniburgh, 2002). Two wells located near the Stanislaus and Mokelumne Rivers differed from other sampled wells and may have a different hydrologic and geochemical history than the other samples with respect to arsenic.

Oxic ground water does not persist within the flow system, and arsenic concentrations in reduced water from wells ranged from 3 to 63 µg/L, with a median concentration of 10 µg/L (fig. 5). Increases in arsenic concentrations under reducing conditions are consistent with reductive dissolution of iron and manganese hydroxide coatings on mineral grains (Smedley and Kinniburgh, 2002). Arsenic in reduced ground water within the study area was positively correlated
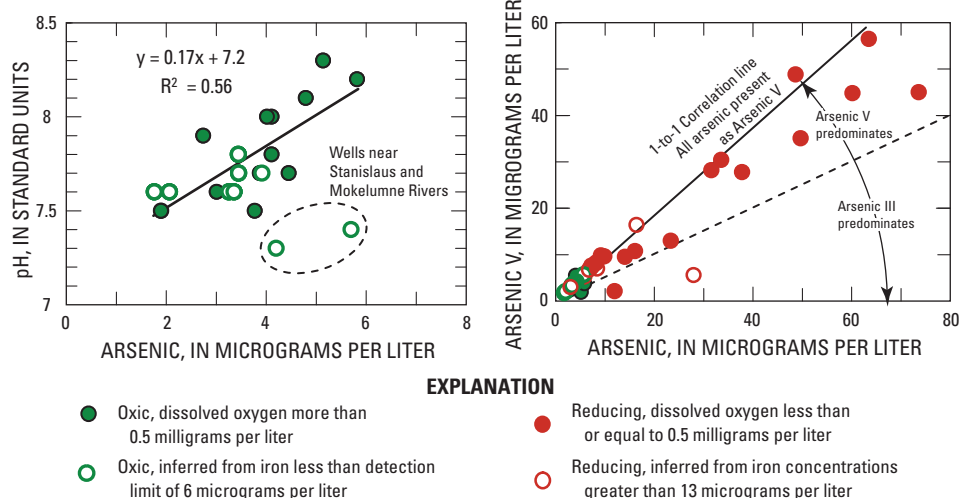
with iron and manganese. The increase in iron and manganese concentrations with increasing arsenic concentration

Figure 5. Arsenic V as a function of arsenic (total dissolved), and pH as a function of arsenic (total dissolved) in water from wells, Eastern San Joaquin Groundwater Subbasin, California, 2006.

were less than would be expected given the abundance of these elements in the environment and suggests that iron and manganese may be subsequently precipitated and removed from solution.

The oxidized form of arsenic, As(V), predominated in water from the surface discharge of almost all sampled wells regardless of pH or redox status (fig. 5). Although As(III) concentrations in some wells were as high as 22 µg/L, only two wells near the San Joaquin Delta had As(III) concentrations greater than As(V) concentrations. Depth-dependent water-chemistry data from well 2N/6E-12J1 shows that the fraction of As(III) increased with depth, as pH increased and dissolved oxygen concentrations decreased, until As(III) was the predominate form (fig. 6).

Microbially mediated reduction of As(V) to As(III) may occur within the ground-water-flow system as oxygen and other electron acceptors are consumed and increasingly anaerobic conditions predominate. In laboratory experiments the reduction of As(V) to As(III) proceeds rapidly compared to sterile (heat-killed) controls (fig. 7). However, the predominance of As (V) on exchanges sites and in ground water suggests that reduction of As(V) to As(III) is slow under natural conditions within the study area.
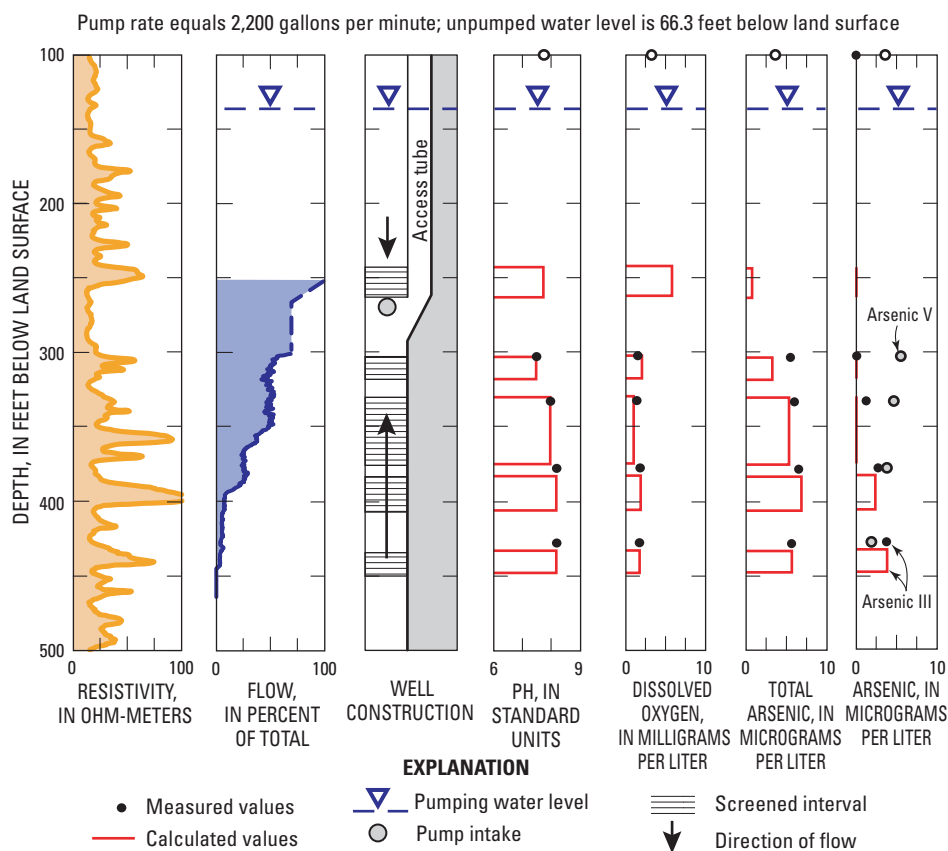

Figure 6. Well-bore flow and depth-dependent water-quality data (including total arsenic, arsenic III, and arsenic V) concentrations from well 2N/6E-12J1 in the Eastern San Joaquin Ground-Water Subbasin, California, August 2006.
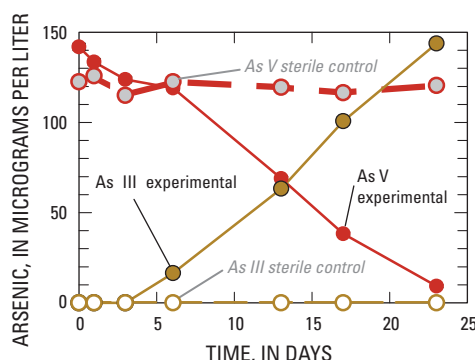
Figure 7. Arsenic III and arsenic V concentrations with time of incubation in a slurry of core material, Eastern San Joaquin Ground-Water Subbasin, California.

Management of Arsenic Concentrations

Two commonly used methods to reduce high-arsenic concentrations in water from public-supply wells are treatment at the surface discharge, or blending of high-arsenic water with water from another source having lower arsenic concentrations. Another potentially cost effective method is modification of the well to exclude water from depths having undesirable concentrations of arsenic (or other constituents).

Well modification was used to lower arsenic concentrations from well 1N/7E- 20N1. This public-supply well had a history of arsenic concentrations above the MCL of 10 µg/L. Well-bore flow and depth-dependent water-quality data show that the high-arsenic concentrations in the surface discharge of the well result from high-arsenic water entering the well from aquifer deposits below about 280 ft (fig. 4). Modification of well 20N1 to eliminate the three deepest well screens and exclude water from the deeper deposits was expected to decrease concentrations in the surface discharge of the well to about 5 µg/L—similar to the arsenic concentration in water from the shallower deposits above 280 ft. Well-bore flow data suggest this would be accompanied by a corresponding decrease in well yield of about 45 percent.

In May 2006, well 20N1 was modified by filling the part of the well below 280 ft with low-permeability bentonite grout, thus sealing the bottom three well screens. A cap of sand was placed on top of the grout to complete the seal. The gravel pack, which extends the length of well 20N1 from the surface seal to 400 ft below land surface, was not altered. After modification, well 20N1 yielded water having an arsenic concentration of 7 µg/L. This concentration was below the MCL, but higher than the concentration of 5 µg/L in aquifer deposits above 280 ft. It is likely that water from deeper depths having high arsenic concentrations moved upward through the aquifer toward the remaining well screens. A radial ground-water flow model with particle tracking was constructed to simulate the movement of water in response to pumping from well 20N1. The model was used to evaluate measured changes in arsenic concentrations resulting from changes in well construction and subsequent upward movement of high-arsenic water from deeper depths.

Model development

The computer program MODFLOW (Harbaugh and McDonald, 1996) was used to simulate the response to pumping from well 20N1. The two-dimensional, radial model consisted of one layer representing a cylinder of aquifer material having a radius of 200,000 ft and a thickness of 400 ft (fig. 8). The radial extent of the model was larger than the influence of simulated pumping from the well. Regional ground-water flow and regional pumping effects were not simulated.

Hydraulic properties representing aquifer materials were assigned from lithologic and geophysical logs. Aquifer materials were assumed to be flat-lying and areally extensive throughout the cylinder. The model was calibrated by adjusting, within reasonable ranges, hydraulic conductivity values associated with the lithology assigned to individual model rows until there was a reasonable match between the simulated and measured distribution of flow within well 20N1, and between the simulated and measured drawdown.

The computer program MODPATH (Pollock, 1994) was used to simulate the movement of water particles within the
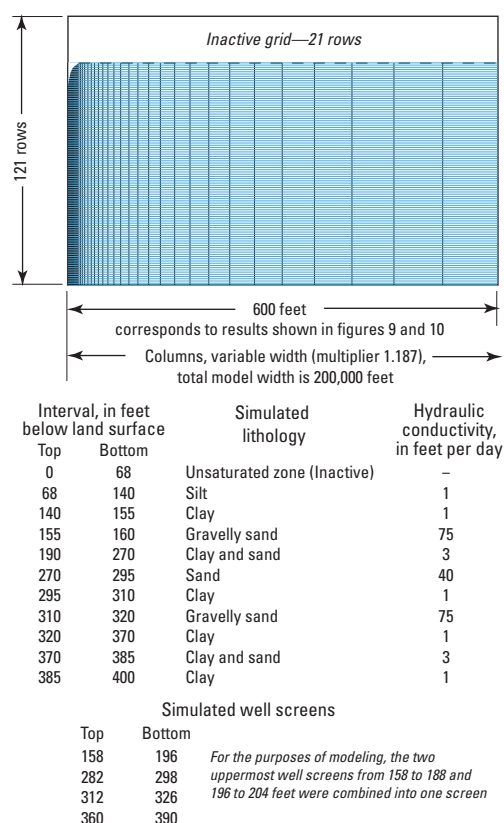

Interval, in feet below land surface		Simulated lithology	Hydraulic conductivity, in feet per day
Top	Bottom		
0	68	Unsaturated zone (Inactive)	–
68	140	Silt	1
140	155	Clay	1
155	160	Gravelly sand	75
190	270	Clay and sand	3
270	295	Sand	40
295	310	Clay	1
310	320	Gravelly sand	75
320	370	Clay	1
370	385	Clay and sand	3
385	400	Clay	1

Simulated well screens		
Top	Bottom	
158	196	*For the purposes of modeling, the two*
282	298	*uppermost well screens from 158 to 188 and*
312	326	*196 to 204 feet were combined into one screen*
360	390	

Figure 8. Model grid, lithology, and well-screen intervals used to simulate flow to well 1N/7E-20N1, Eastern San Joaquin Ground-Water Subbasin, California.

model domain. Each particle represents a discrete fraction of the well yield and a unique quality of water. The quality of water yielded by well 20N1 was calculated as the flow-weighted average of the particle concentrations.

Simulated well construction was changed to determine the effect of modifying the well on arsenic concentrations in the surface discharge of water from well 20N1. Simulated results were compared to measured changes in arsenic concentration in water from the surface discharge of the well after the three deepest screens were backfilled with low-permeability bentonite grout.

Additional details concerning model construction are available in Table 1.

Model results

Pumping was simulated at the rate of 2,300 gal/min for 1,000 days, and the model simulated drawdown of 58 ft closely approximated the measured drawdown of 61 ft. Simulated drawdown

from pumping by the unmodified well was distributed throughout the aquifer (fig. 9). The simulated movement of water (illustrated as particles) to the well was primarily through coarse-grained deposits with smaller contributions from the intervening finer-grain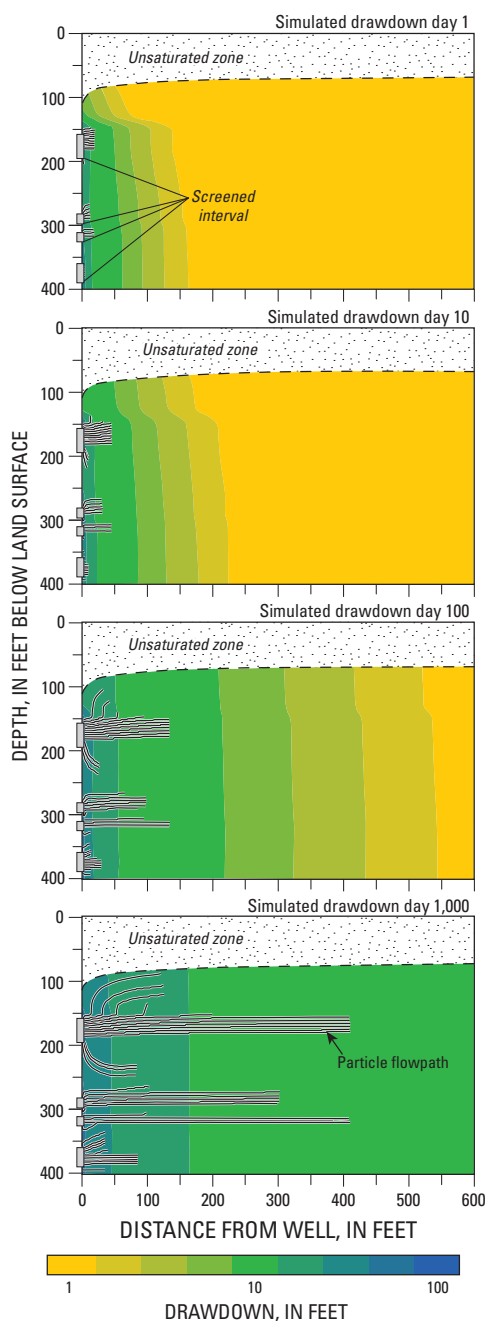ed deposits. Simulations show that near the water table, water moved almost vertically downward until it encountered a coarse-grained layer and then moved rapidly towards the well. Finer-grained deposits in the deeper parts of the aquifer within 500 ft of the well were still contributing water (particles) after 1,000 days of simulated pumping.

Pumping from the modified well was simulated at a rate 1,600 gal/min for 1,000 days. The lower pumping rate was required to maintain water levels above the well screens, and reflects the expected lower yield from the modified well. Simulated drawdown from the modified well was focused at the upper screened interval (fig. 10). Water from deeper depths no longer entered the well directly, but instead moved upward through the aquifer deposits before entering the well. During the 1,000 day simulation, ground water from the deeper deposits contributed increasingly to the yield of the well, reaching a maximum of about 20 percent, increasing arsenic concentrations during the simulation (fig. 11). However, the volume of high-arsenic water yielded from the deeper deposits was less than the 45 percent of the volume of water yielded from those deposits prior to modification of the well.

The model was used to calculate the change in arsenic concentration in the surface discharge of well 20N1 if different screened intervals were backfilled with grout (fig. 11). The small change in the arsenic concentration as a result of filling the deepest screen interval was expected because of the small yield from that depth. Larger changes in arsenic concentrations were simulated as a result of the filling of the second and finally the third deepest screen intervals. Simulated arsenic concentrations were lower initially, but increased with time as water from deeper parts of the aquifer moved upward in response to pumping. The simulated arsenic concentration in the surface discharge of well 20N1 after the three deepest screens were filled with grout ultimately increased to a concentration of 7.2 µg/L after 1,000 days of pumping (fig. 11).

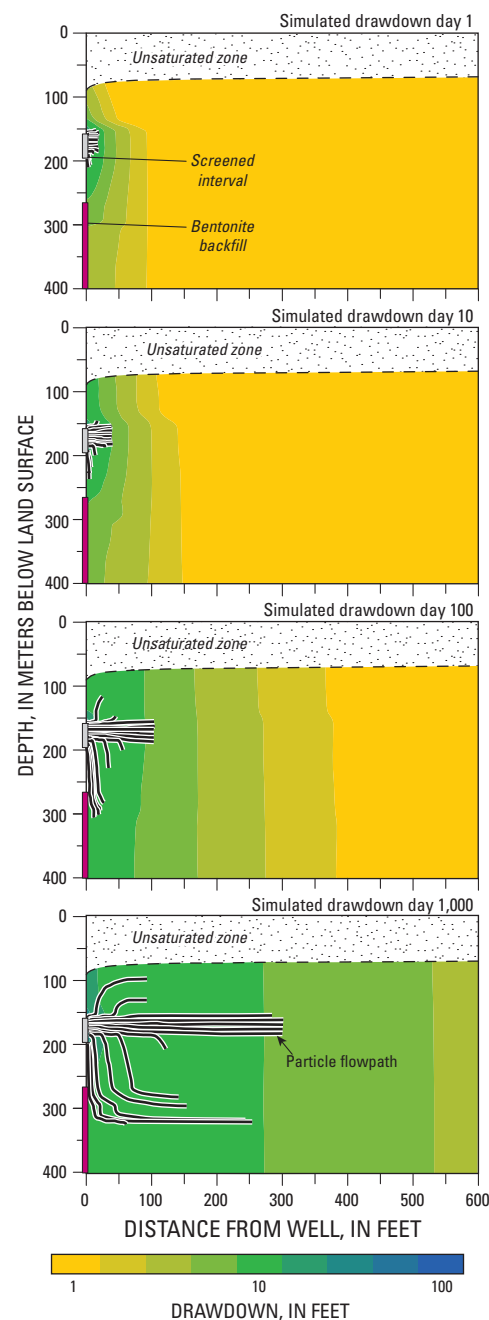Simulated sealing of the gravel pack produced results that were only slightly different from results shown in

Figure 9. Simulated pressure response and particle movement during pumping prior to modification of well 1N/7E-20N1, Eastern San Joaquin Ground-Water Subbasin, California.

Figure 10. Simulated pressure response and particle movement during pumping after modification of well 1N/7E-20N1, Eastern San Joaquin Ground-Water Subbasin, California.

figure 11. Despite its high-permeability, the gravel-packed packed annulus is not large enough to allow the movement of large quantities of high-arsenic water to the upper screen when the modified well is pumped. This suggests that relatively simple well modification strategies that only require backfilling the well and not
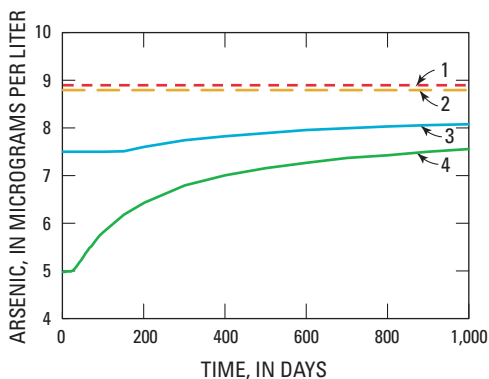
Simulated well screens—
1. 158 to 196 feet, 282 to 298 feet, 312 to 326 feet, 360 to 390 feet (original well construction)
2. 158 to 196 feet, 282 to 298 feet, 312 to 326 feet
3. 158 to 196 feet, 282 to 298 feet
4. 158 to 196 feet (modified well construction)

Figure 11. Simulated arsenic concentrations as a function of time after modification of well 1N/7E-20N1, Eastern San Joaquin Ground-Water Subbasin, California.

the destruction of the well, casing, and surrounding gravel pack can be used to manage arsenic concentrations in water from wells.

Model Limitations

The two-dimensional radial flow model developed to interpret well-bore-flow and depth-dependent water-quality data from well 1N/7E-20N1 is a simplified representation of the flow system around the well. The flat-lying areally extensive aquifer materials are not intended to accurately represent regional changes in subsurface geology, including the areal extent of these materials and the hydraulic connection between those materials. Similarly, the flat water table does not represent regional ground-water flow or interactions between pumping wells. However, this simplified radial-flow model does provide a simple tool to evaluate the effects of changing well design on the quality of water that is discharged from a well.

Acknowledgements

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