U.S. Department of the Interior
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RELATION OF MERCURY TO OTHER CHEMICAL CONSTITUENTS IN GROUND WATER IN THE KIRKWOOD-COHANSEY AQUIFER SYSTEM, NEW JERSEY COASTAL PLAIN, AND MECHANISMS FOR MOBILIZATION OF MERCURY FROM SEDIMENTS TO GROUND WATER

Water-Resources Investigations Report 00-4230

Prepared in cooperation with the

NEW JERSEY DEPARTMENT OF ENVIRONMENTAL PROTECTION

USGS
science for a changing world

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U.S. GEOLOGICAL SURVEY

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U.S. DEPARTMENT OF THE INTERIOR

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CONTENTS

Page

CONTENTS--Continued

Page

ILLUSTRATIONS

ILLUSTRATIONS--Continued

Page

TABLES

CONVERSION FACTORS, VERTICAL DATUM, AND ABBREVIATIONS

Multiply	By	To obtain
	Length	
inch (in.)	25.4	millimeter (mm)
inch (in.)	2.54	centimeter (cm)
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer
	Area	
acre	4,047	square meter (m^2)
acre	0.4047	hectare
square foot (ft^2)	0.09294	square meter
square mile (mi^2)	2.590	square kilometer
	Volume	
ounce, fluid (fl. oz)	29.57	milliliter (mL)
ounce, fluid (fl. oz)	0.02957	liter (L)
gallon (gal)	3.785	liter
	Mass	
ounce, avoirdupois (oz)	28.35	gram (g)
ounce, avoirdupois (oz)	28,349	milligram (mg)
ounce, avoirdupois (oz)	28,349,000	microgram (µg)
pound, avoirdupois (lb)	0.4536	kilogram (kg)
	Temperature	
degree Fahrenheit (°F)	°C = 5/9 x (°F-32)	degree Celsius (°C)

Vertical Datum

Vertical datum: In this report "sea level" refers to the National Geodetic Vertical Datum of 1929-- a geodetic datum derived from a general adjustment of the first-order level nets of the United States and Canada, formerly called Sea Level Datum of 1929.

Mass, length, water-quality, soil-concentration, load, and chemical abbreviations

ng - nanogram
nm - nanometer
µm - micrometer
ng/L - nanograms per liter (parts per trillion)
µg/L - micrograms per liter (parts per billion or ppb)
mg/L - milligrams per liter (parts per million or ppm)
µg/kg - micrograms per kilogram (parts per billion)
µg/g - micrograms per gram (parts per million)
mg/m^3 - milligrams per cubic meter
kg/ha - kilograms per hectare
M - Molar, or moles
µM - micromolar, or micromoles
mV - millivolts
K_H - dissociation constant
N - nitrogen
K - potassium
P - phosphorus

RELATION OF MERCURY TO OTHER CHEMICAL CONSTITUENTS IN GROUND WATER IN THE KIRKWOOD-COHANSEY AQUIFER SYSTEM, NEW JERSEY COASTAL PLAIN, AND MECHANISMS FOR MOBILIZATION OF MERCURY FROM SEDIMENTS TO GROUND WATER

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ABSTRACT

Water from 265 domestic wells that tap the unconfined Kirkwood-Cohansey aquifer system in the Coastal Plain of New Jersey contained concentrations of mercury that are equal to or exceed the U.S. Environmental Protection Agency maximum contaminant level (MCL) of 2 µg/L (micrograms per liter). The wells range in depth from 50 to 200 feet, and are located in 32 discrete, mostly residential, areas that were developed primarily on former agricultural land during the 1950's through the 1970's. Concentrations in two other areas exceeded 1 µg/L. Naturally occurring mercury concentrations in ground water from the Kirkwood-Cohansey aquifer system typically are less than 0.01 µg/L, but concentrations in water from some wells were as much as 42 µg/L. No evidence currently exists that conclusively links known point sources such as landfills, industrial operations, or commercial enterprises to most of the elevated concentrations of mercury in ground water in the residential areas. Possible sources of the mercury include pesticides and atmospheric deposition.

Analysis of water from wells in 6 of the 34 areas for other constituents indicates that nitrate concentrations also commonly are elevated above background levels (which typically are undetectable at 0.01 milligrams per liter), and exceed the MCL of 10 milligrams per liter in some samples. Several volatile organic compounds (VOCs), including chloroform, also have been measured in water from wells at many of the 34 sites. Analytical results for water samples collected at several depths from boreholes at 2 of the 34 sites indicate elevated concentrations of calcium, magnesium, barium, strontium, nitrate, and chloride, which may be related to both agricultural chemical applications and septic-system effluent. Determinations of tritium and helium concentrations indicate that water containing elevated concentrations of mercury recharged the aquifer between 9.4 and 79 years ago, which includes the period during which many of the 34 sites were undergoing a change from agricultural or undeveloped to residential land use.

Batch equilibrium experiments were used to measure adsorption of dissolved mercury, mercuric chloride, and phenylmercuric acetate by aquifer sediments at pH 3.5-4.0, 4.5-5.0, and 5.5-6.0. In nearly all cases, 55 to 95 percent of the mercury added to the sediments was adsorbed. Mercury mobilization from aquifer sediments inoculated with mercury was investigated by leaching the sediments with two concentrations of nitric acid (a component of acid rain), a sodium chloride solution (simulating road salt), and three fertilizer solutions. A solution of 20-20-20 (nitrogen-phosphorous-potassium) fertilizer removed virtually all of the mercury with which the sediments had been inoculated. The sodium chloride solution was moderately effective in removing applied mercury from the sediments, as was a solution of nitric acid. A more dilute nitric acid solution and two sodium nitrate fertilizer solutions were less effective.

Results of these experiments indicate that mercury adsorbs to aquifer sediments, but that varying amounts can be removed by infiltrating solutions, some of which can be related to specific

land uses. Land-use history at the 34 sites generally indicates a change from agricultural or undeveloped settings to residential settings. Whatever the source of mercury to these sites, a change in the geochemical environment of the soil and aquifer brought about by land-use change probably provides mechanisms for mobilizing the mercury from soils and sediments to ground water.

INTRODUCTION

The discovery of elevated concentrations of mercury in ground water in excess of the U.S. Environmental Protection Agency (USEPA) maximum contaminant level (MCL) of 2 μg/L (U.S. Environmental Protection Agency, 1994) in southern New Jersey prompted a study to compile available mercury-concentration and other water-quality data and well-construction data for wells tapping the affected aquifer, to assess local hydrology, and to investigate possible point sources of mercury contamination (Barringer and others, 1995; Barringer and others, 1997). Mercury-concentration data for water from 2,270 wells tapping the areally extensive, unconfined Kirkwood-Cohansey aquifer system in southern New Jersey were collected by State, county, and local agencies during 1982-92; 2,239 of these wells were located in 34 discrete areas where one or more wells yielded water containing mercury at concentrations greater than 1 μg/L. The wells range in depth from 50 to 200 ft, and are, in nearly all instances, located in residential areas. Of these wells, 306 yielded water at least once that contained mercury at concentrations that exceeded the MCL, and 265 at 32 areas yielded water containing mercury at concentrations above the MCL at the most recent sampling. About 1,000 wells yielded water containing mercury at concentrations between the reporting limit and 2 μg/L; these concentrations, although below the MCL, are higher than those that have been determined to occur naturally in the aquifer (Murphy and others, 1994). Not only are aquifer sediments unlikely to be the source of elevated (greater than 1 μg/L) mercury concentrations in ground water (Dooley, 1992), but results of a previous investigation showed that potential point sources such as landfills, industrial operations, or commercial enterprises could not be linked conclusively to the 34 sites (Barringer and others, 1997). Use of mercurial pesticides on crops and lawns appears to be one likely source of elevated concentrations of mercury in ground water (Murphy and others, 1994; Barringer and others, 1997).

Water-quality data other than mercury analyses are relatively sparse for ground water from the affected residential areas, but concentrations of chloride and (or) nitrate that are larger than background concentrations are reported for 6 of the 34 sites where elevated mercury concentrations are present. Additionally, several volatile organic compounds (VOCs), such as chlorinated solvents and chloroform, have been detected in water from some of the wells in the residential neighborhoods, although water samples containing elevated mercury concentrations seldom contain elevated concentrations of VOCs. Therefore, in order to determine how the mercury has moved from the land surface or shallow subsurface to ground water, the U.S. Geological Survey (USGS), in cooperation with the New Jersey Department of Environmental Protection (NJDEP), began a study to identify (1) other chemical constituents indicative of particular anthropogenic sources, and (2) possible agents of mobilization or geochemical processes associated with residential and agricultural settings.

Purpose and Scope

This report describes the relations between mercury and other chemical constituents in ground water that were determined on the basis of analysis of newly collected water samples and analysis of available data for six sites of mercury-contaminated ground water, and by sampling ground water at different depths at two of the sites. It also identifies possible mobilizing agents determined from results of experiments in which adsorption and desorption of mercury to and from aquifer sediments were measured.

Mercury in the Environment

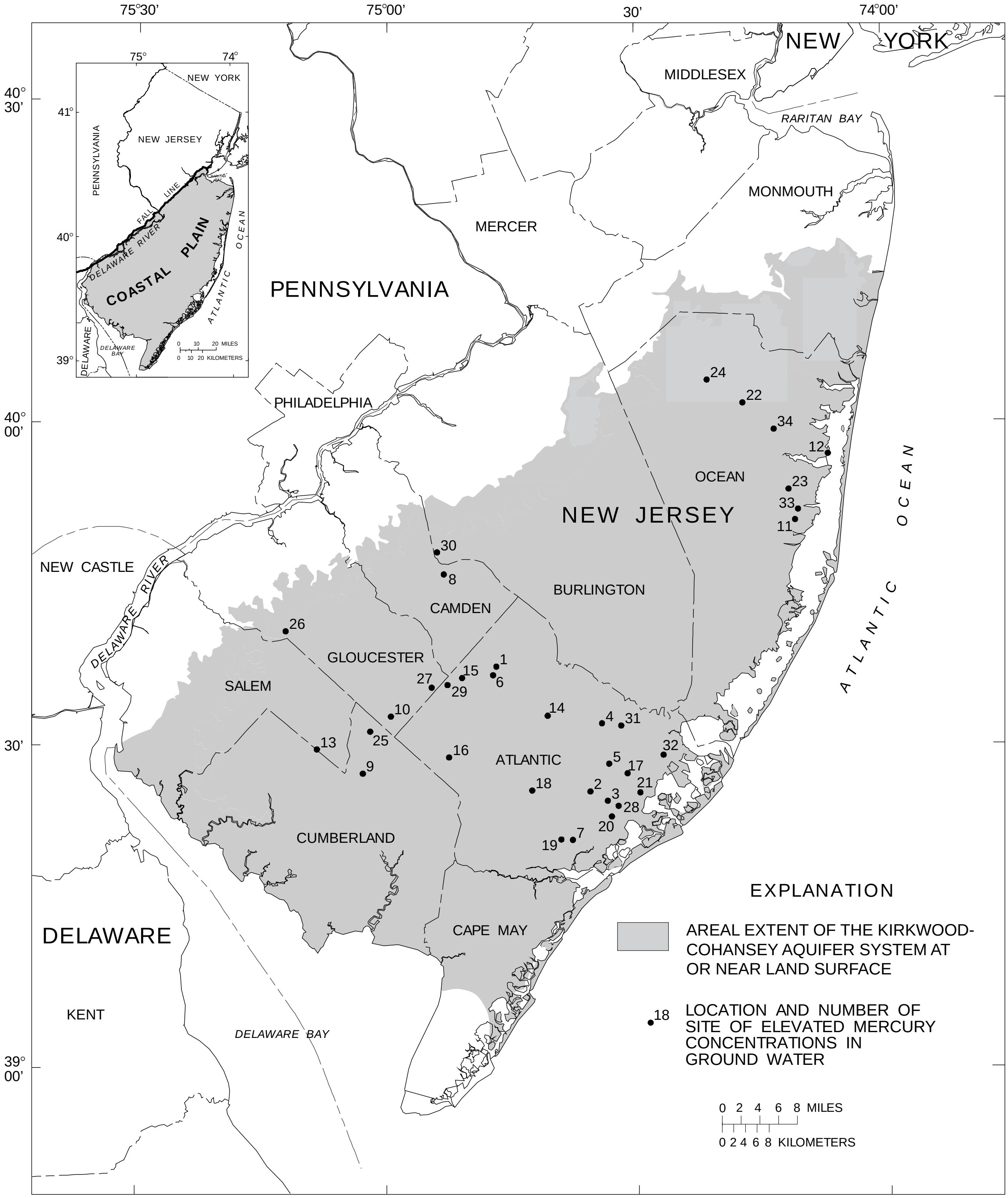
Many measurements of mercury in the environment have been directed at airborne contaminants (for example, Brosset, 1982; Morrison and others, 1995) and, in the aquatic environment, studies have focused almost entirely on surface-water systems. There are many reports of mercury-contaminated freshwater fish (for example, Snarski and Olsen, 1982; Boudou and Ribeyre, 1984; Niimi and Jowe-Linde, 1984; Sorenson and others, 1990), including fish in New Jersey (Academy of Natural Sciences, 1994). Mercury concentrations in lake sediments and water have been assessed (for example: Anderson and others, 1987; Fitzgerald and Watras, 1989; Glass and others, 1991; Lee and Hultberg, 1990; Swain and others, 1992). Relatively few instances of elevated concentrations of mercury in ground water have been reported, except when mercury was included among other contaminants in site-specific hazardous-waste investigations (for example, Ebasco, 1991), or other investigations of ground-water contamination in industrialized areas (for example, Srikanth and others, 1993; Somasundaram and others, 1993). In a recent study, Sidle (1993) attributes elevated concentrations of mercury in shallow ground water in Maine to leaching of the metal from the granitic aquifer. Other studies of mercury in ground water in Sweden (Lindqvist and others, 1991), Wisconsin (Krabbenhoft and Babiarz, 1992), and New Jersey (Windom and Smith, 1992) have shown that ambient mercury concentrations generally are less than 15 ng/L. Thus, given that elevated mercury concentrations in ground water typically appear to be associated with hazardous-waste sites or geologic sources, it is not known whether the elevated concentrations that have been measured in ground water in southern New Jersey are representative of a more widespread problem that is yet to be discovered elsewhere, or whether they represent an isolated phenomenon.

History of Mercury Contamination in the Kirkwood-Cohansey Aquifer System

Instances of mercury concentrations in excess of the MCL in water from domestic wells that tap the Kirkwood-Cohansey aquifer system in southern New Jersey were reported as early as 1982 (unpublished records on file at N.J. Department of Environmental Protection, Trenton, N.J.). This aquifer system is the most areally extensive unconfined (water-table) aquifer in the Coastal Plain of New Jersey and is a major source of water for both domestic and public-supply wells. During the 1980's, the number of instances of mercury-contaminated ground water increased, but as a result of the sporadic nature of the early reports, the contamination was not perceived as a widespread problem until 1988, when State and county agencies began sampling and analyzing ground water more frequently. Because most of the affected wells were in residential areas, many of which are post-World War II housing developments, the water-quality data collected by State and county agencies tend to be clustered, as response to an instance of mercury contamination prompted sampling of the surrounding wells in a given neighborhood.

Concentrations

In 1992, the USGS began a 2-year study, in cooperation with the NJDEP, to determine the extent of the mercury-contaminated ground water in the New Jersey Coastal Plain, and to advance and test hypotheses as to its sources and mobility. On the basis of data for a total of 2,270 wells compiled from State and county sources, 2,239 wells were found to be associated with 34 discrete areas ("sites") in which one or more wells at each site had yielded water with mercury concentrations of 1 µg/L or greater (Barringer and others, 1997) (fig. 1)--well above naturally occurring concentrations (Windom and Smith, 1992; Murphy and others, 1994), and therefore are considered to represent anthropogenic inputs of mercury. Of the 2,239 wells at the 34 sites, 306 yielded water with mercury concentrations that had exceeded the MCL on at least one sampling occasion; results of the most recent sampling at each well indicate that 265 of the 2,239 wells yielded water with mercury concentrations exceeding the MCL (Barringer and others, 1997). Recently measured concentrations were as much as 42 µg/L, and 35 wells yielded water with concentrations greater than 10 µg/L. Mercury was not detected in water from 927 of the 2,239 wells. Detectable mercury concentrations at or less than the MCL were measured in water from 1,047 of the 2,239 wells. If only the most recent data for each well are considered, the median mercury con-



Base modified from U.S. Geological Survey digital
data, 1:100,000, 1983, Universal Transverse
Mercator projection, Zone 18

Figure 1. Locations of 34 sites of elevated mercury concentrations in ground water, and the areal extent of the unconfined Kirkwood-Cohansey aquifer system, New Jersey Coastal Plain. (Inset map shows location of the New Jersey Coastal Plain)

centration in samples from the 2,239 wells was less than 1 µg/L (Barringer and others, 1997).

Recently measured mercury concentrations above the MCL, reported subsequent to the sampling, analysis, and experimental results reported here, are present at several additional sites in Ocean and Atlantic Counties. Investigation of these sites by NJDEP currently (2000) is ongoing. Additional samples recently collected by the USGS from six public-supply wells contained mercury at concentrations between the reporting limit (0.1 µg/L) and the MCL (Ivahnenko and others, 1996). These data indicate that the full extent of the instances of elevated mercury concentrations has yet to be defined.

Well-depth data were available for only 20 percent of the wells at the 34 sites identified before or during the USGS study. Concentrations of mercury commonly were largest at depths from 50 to 120 ft below land surface, but the apparent vertical distribution probably is, in part, a function of the depths to which domestic wells typically are installed (Barringer and others, 1997). Data for 13 wells less than 50 ft in depth and data from incremental borings at 2 of the 34 sites investigated in this study indicate that, to date (1998), mercury detections in shallow (less than 50 ft deep) ground water at concentrations greater than 0.1 µg/L (the lowest reporting limit for the laboratories analyzing the water samples) apparently are relatively uncommon, but the number of shallow wells for which mercury data are available is very small relative to the number of wells in the data set.

Possible Sources

The source of elevated mercury in ground water from the Kirkwood-Cohansey aquifer system is not natural (Dooley, 1992). Mercury concentrations in the clay lenses that are found throughout the aquifer include a concentration of 108 µg/kg in a clay lens from a core at a site of elevated concentrations of mercury in ground water (site 6; see fig. 1) and a concentration of about 40 µg/kg in a red silt-clay layer from another site (site 10; see fig. 1). The mercury in the clays and silts is presumed to occur naturally. The rate of release of mercury from clay lenses probably is low because mercury binds strongly to clay, and the passage of water through clays is slow relative to that through the surrounding sands. Therefore, leaching of mercury from clay lenses probably does not account for the elevated concentrations of mercury measured in ground water in the study area. Naturally occurring mercury concentrations in ground water in the Kirkwood-Cohansey aquifer system are less than 10 ng/L (0.01 µg/L) (Windom and Smith, 1992; Murphy and others, 1994).

The amount of mercury that could have been deposited on the land surface from the atmosphere is estimated to be fairly substantial, but measurements of mercury in undisturbed forest soils in several areas of the Coastal Plain indicate that much of atmospherically deposited mercury is sequestered in organic-rich soil horizons (Barringer and others, 1997; Barringer and others, 1998). Outgassing of mercury from exterior house paint in residential areas is likely to contribute to atmospheric levels (Barringer and others, 1997), but no data are available to evaluate the transport, distribution, or concentration of the mercury vapor, or the possibility of mercury leaching from painted surfaces.

Because about 75 percent of the housing developments where concentrations of mercury in ground water are elevated were built on former agricultural land, mercurial pesticides, known to have been used in New Jersey, represent one possible source of the mercury. Mercuric chloride and mercurous chloride were used on some row crops in the United States from the end of the 19th century until the mid-20th century. During the 1950’s, organomercurials such as phenylmercuric acetate (PMA) were preferred, replacing the inorganic pesticides. PMA was used as a fungicide on turf and fruit trees until 1972, when its use on food crops was banned (D’Itri, 1972). PMA is still used on golf courses in the United States, although it is no longer registered for use in New Jersey. Various mercurial pesticides also were used as seed dressings, primarily on corn. The actual amount of mercurial pesticides applied in southern New Jersey is not known, but, if recommended practices were followed, past use of the pesticides in agriculture and on turf, including residential lawns, could represent a substantial source of mercury. Some fertilizers contain substantial amounts of mercury (Andersson, 1979). Additionally, improper disposal of mercurial wastes in unpermitted dumps could create point sources of contamination, but locations of such dumps generally are not known.

The wells yielding water with elevated concentrations of mercury generally are screened at depths ranging from 50 to 200 ft. On the basis of age-dating measurements and simulated travel times in the Kirkwood-Cohansey aquifer system (Rice and Szabo, 1997), the contaminated water is probably more than a decade old. The deeper wells tap water that may have been recharged from the land surface about 50 or more years ago. Investigations of local hydrology, past land use, and known point sources of contamination led to the conclusion that, for most of the residential neighborhoods in which concentrations of mercury in ground water were elevated, point sources such as landfills, military installations, industrial operations, and commercial enterprises were unlikely to have contributed mercury to the ground water underlying the housing developments (Barringer and others, 1997).

Acknowledgments

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CHEMISTRY AND MOBILITY OF MERCURY AND COLLOIDAL TRANSPORT

Mercury chemistry is complex, and the element can be involved in a wide variety of reactions. Because mercury binds strongly to materials that can become colloids, mercury mobility may be controlled not only by chemical reactions, but by chemical environments that promote mobility of colloids.

Chemistry and Mobility of Mercury

In the terrestrial environment, mercury can be present in three oxidation states: the metal (Hg^0) and two ionized forms, mercurous ion (Hg_2^{2+}) or Hg(I), and mercuric ion (Hg^{2+}) or Hg(II). Hg^0, with a vapor pressure of 14 mg/m^3 at 20 °C (Andren and Nriagu, 1979), is sparingly soluble in distilled, neutral-pH water, giving a concentration of 2.9×10^{-7} M, or 58 µg/L. Mercurous ion is not stable under most pressures. Ionic mercury (Hg(II)) tends to interact covalently with bases such as sulfide, ammonia, and chloride rather than with oxygen-donor ligands (Schuster, 1991).

Ionic mercury forms strong covalent complexes with various organic ligands, including cysteine, amino acids, and hydroxy carboxylic acids (Moore and Ramamoorthy, 1984). Equilibrium complex formation constants (Ks) are large; log Ks at 25 °C for Hg^{2+} with cysteine and glycine are 43.57 and 10.3, respectively (Leckie and James, 1974), although the log K reported by Martell and others (1993) for cysteine is smaller (14.2). Other log Ks reported are for acetate (8.4; Martell and others, 1993) and oxylate (6.98; Lindsay, 1979). Therefore, naturally occurring organic matter tends to complex strongly with mercury, and results of several studies have shown that humic acids bond strongly with mercury, although the mechanisms are not well understood (Thurman, 1985; Biester and Zimmer, 1998). In fact, mercury is retained more strongly by humic acid than 10 other metals at pHs from 2.4 to 5.8 (Kerndorff and Schnitzer, 1980).

Humic substances also may play a role in reducing ionic mercury to the elemental form in aqueous systems (Allard and Arsenie, 1991). Additionally, soil organic matter appears to reduce ionic mercury applied to soils as salts ($HgCl_2$, $Hg(NO_3)$, and Hg (OCH_3COO_2), resulting in the apparent evolution of vaporous Hg^0 (Frear and Dills, 1967). The same phenomenon has been suggested in a study of mercury salts applied to turf grass (Gilmour and Miller, 1973).

Ionic mercury (Hg^{2+}) is subject to methylation, needing only the presence of methyl donor molecules and a biological and (or) chemical environment that can promote the process. Investigations of mercury cycling in lakes indicate that methylation of mercury is primarily biologically mediated, chiefly under anaerobic conditions. Gilmour and others (1992) found methylmercury production highest in lake sediments near the sediment-water interface, and concluded that sulfate-reducing bacteria mediated the methylation reaction. Zillioux and others (1993), reviewing several methylation studies, conclude that methylation of mercury is related to fermentation occurring during sulfate reduction rather than to sulfate reduction alone. Methylation can also occur abiotically, and in aerobic systems as well (Gavis and Ferguson, 1972; Schroeder and others, 1989). In a study of mercury methylation in agricultural soils amended with mercuric nitrate, concentrations of methylmercury were larger in sterilized (autoclaved) soils than in unsterilized soils (Rogers, 1976).

The retention and distribution of mercury in soils and sediments have been investigated for nearly 3 decades (for example: Lockwood and Chen, 1973; Reimers and Krenkel, 1974; Hogg and others, 1978, Kinniburgh and Jackson, 1978; Amacher and others, 1986; Amacher and others, 1990; Schuster, 1991). Many of these studies have shown that sediments with high organic content have a high adsorption capacity for mercury and that oxides of iron and manganese also can provide adsorption sites for mercury. In this respect, the chemical behavior of mercury appears similar to that of several of the other heavy metals, such as lead and zinc.

There are some important differences between mercury and other heavy metals, however. Metal ions in solution tend to be hydrated; that is, they acquire a shell of water molecules. If one of the octahedrally coordinated water molecules dissociates into hydrogen ion (H^+) and hydroxyl ion (OH^-), the result is a reactive hydrated metal ion (Barrow and Cox, 1992). For most metals, this dissociation occurs at pHs greater than 8, but for mercury (as Hg(II)), the dissociation takes place at acidic pHs (Baes and Mesmer, 1976). In a solution of $HgCl_2$, in which mercury and chloride ions are completely dissociated, the hydrolysis of mercury produces hydrogen ions through the reactions

$$Hg^{2+} + H_2O = HgOH^+ + H^+, \quad K_{H1} = 10^{-3.7}, \text{ and}$$

$$HgOH^+ + H_2O = Hg(OH)_2{}^o + H^+, \quad K_{H2} = 10^{-2.6}$$

(Pankow, 1991, p. 366).

Mercury in solution occurs as $Hg(OH)^+$ at low pHs; with increasing pH, the concentration of the uncharged species $Hg(OH)_2{}^o$ increases. Both species contain mercury as Hg(II).

In acid solutions, adsorption sites on solid phases (oxides, or clays, for example) are protonated--that is, they are populated with positively charged hydrogen ions; thus, adsorption of negatively charged species in solution is favored. Adsorption of positively charged species becomes increasingly favored as pH increases. For the hydrated mercury ion ($Hg(OH)^+$), adsorption increases as pH increases up to about pH 4; above pH 4, the concentration of $Hg(OH)^+$ decreases as $Hg(OH)_2{}^o$ forms, and, despite the increasingly favorable potential of a solid for adsorption of positively charged species, adsorption of mercury decreases slightly (Barrow and Cox, 1992).

In natural aqueous systems with low chloride-ion concentrations, mercury hydroxide complexes bind to surfaces of aluminum, iron, and manganese oxides, hydroxides, and oxyhydroxides. Even neutral mercury hydroxide species can adsorb to oxide surfaces by the hydroxide bridges (Rai and Zachara, 1984). In chloride-rich waters, however, mercury forms strong complexes with chloride at pHs from about 2 to 6 (Barrow and Cox, 1992). Mercury chloride species are favored at low pH (less than 4); an intermediate species, HgClOH, is postulated between about pH 4 and pH 4.5; and $Hg(OH)_2$ becomes significant as pH increases (Kinniburgh and Jackson, 1978). In experiments involving mercury binding to soils, mercury retention decreased dramatically in the presence of chloride, as mercury chloride species were formed (Andersson, 1970).

Chloride ion, present over a wide range of concentrations, decreases the adsorption of mercury (Hg(II)) to goethite, silica, and iron hydrous oxide gel, and the humic acid fraction of organic matter (Forbes and others, 1974; MacNaughton and James, 1974; Kinniburgh and Jackson, 1978; Lumsdon and others, 1995). In experiments in which mercury is adsorbed to goethite, increasing chloride concentrations from 5 to 5,000 μM (0.18-177 mg/L) decreased mercury adsorption at pHs less than 4, and increased the pH at which maximum adsorption occurred (Barrow and Cox, 1992). Solutions containing chloride concentrations similar to those in landfill leachate (0.1 M in chloride ion or 3,545 mg/L chloride) suppressed mercury adsorption to soils containing clays, organic matter, and hydrous iron oxides; over a pH range of 3 to 7, mercury was calculated to be present mostly (75 percent) as the neutral species $HgCl_2$, with the remaining 25 percent being made up nearly equally of two negatively charged species ($HgCl_3^-$ and $HgCl_4^{2-}$) (Lumsden and others, 1995). Mercury (Hg(II)) adsorption to iron hydrous oxide gel was less (68-70 percent) at pH 4.50 in a solution 1×10^{-4} M in chloride (3.5 mg/L chloride) than in a chloride-free solution (91.5 percent) at the same pH, whereas, at pH 5.95, the percent mercury sorbed was about the same for both types of solution (Kinniburgh and Jackson, 1978). Mercury adsorption to the iron hydrous oxide gel, in a 4×10^{-6} M chloride solution (0.14 mg/L chloride) was found to plateau at about pH 5, which was ascribed to the great stability of $Hg(OH)_2^o$ in solution, and a decrease in adsorption of mercury was found at pH above 6.5 (Kinniburgh and Jackson, 1978).

This chemical behavior of mercury species is anomalous with respect to the adsorption of other heavy metal complexes; in some cases, chloride ion has been found to increase the adsorption of other metals on various hydrous iron oxide substrates. The anomalous behavior may arise from the linear configuration of the mercury chloride complexes, which prevents strong multidentate bonding (Kinniburgh and Jackson, 1978), although, as summarized by Lumsden and others (1995), ternary surface-metal-chloride complexes of mercury, cadmium, and lead form on iron oxide (geothite) surfaces (Gunneriusson, 1994; Gunneriusson and Sjoberg, 1993; Gunneriusson and others, 1994). These complexes do not form on the surfaces of humic substances, however (Lovgren and Sjoberg, 1989).

In addition to chloride complexes, with stability constants of about $K = 10^{15}$ (Moore and Ramamoorthy, 1984), mercury forms other halide complexes. The stability constants given for Hg(II) complexes of iodide and bromide are larger than those for the formation of various chloride complexes (see Lindsay, 1979, p. 346). Mercury (Hg(II)) also forms complexes with nitrate, ammonium, and sulfate ligands, but the stability constants for these complexes are substantially smaller than those for the halide ligands (Lindsay, 1979).

Several mercury compounds have been applied to soils as pesticides in the last century or so; the adsorption of mercury from these pesticides to soils is dependent on the mercury species in solution (Hogg and others, 1978; Inoue and Aomine, 1969). Maximum reported adsorption capacities for mercury on two soils range from 20 μg/g (or parts per million) for methyl mercury chloride to 2,465 μg/g for mercuric chloride (Hogg and others, 1978).

The desorption of mercury from soils and sediments also has been studied. With respect to the effects of runoff on mercury transport, Feick and others (1972), Wang and others (1991), and

Elliott and Linn (1987) found that mercury movement in soils could be facilitated by an increase in chloride-ion concentration. Similar results were obtained in column studies using quartz sand leached with 10^{-2} M sodium chloride; the mercury form in these experiments is reported to be the anion $HgCl_4^{2-}$ (Behra and Bourg, 1987). In an earlier study (Behra, 1986), mercury associated with colloids was released by a solution containing 3.5×10^{-2} M sodium perchlorate and 10^{-2} M sodium chloride. In experiments with mercury-inoculated bay sediments, moderately acid (pH 5), reduced (-150 mV) conditions were found to enhance concentrations of mercury in solution (Gambrell and others, 1980); the dissolved mercury was present mostly in inorganic, nonionic, or negatively charged complexes, or was complexed with soluble organic material. In contaminated coastal sediments, less mercury was present in the solution as ionic strength of the leaching solution increased, presumably because an increase in ionic strength destabilized colloids to which the mercury was bound (Duarte and others, 1991).

Colloidal Transport

Studies of contaminant transport in the subsurface typically have considered partitioning of contaminants between immobile aquifer solids and the mobile aqueous phase, but have infrequently included mobile colloids as a mode of transport (McCarthy and Zachara, 1989). Recent studies of contaminant transport (Gschwend and Reynolds, 1987; Enfield and Bengtsson, 1988; Buddemeier and Hunt, 1988) have shown that the amount of suspended colloids present in ground water can be substantial, and that various contaminants are adsorbed to these colloids. Although the upper limit for the size of particles considered to be colloids has varied over an order of magnitude, colloids generally are defined in recent literature as particles less than 1 μm in diameter (Mills and others, 1991). Colloids can be composed of macromolecular organic carbon, microorganisms, inorganic substances such as weathering products and mineral precipitates, and microemulsions of nonaqueous phase liquids (McCarthy and Zachara, 1989). Colloids composed of hydrous iron and aluminum oxides or clay minerals, in particular, can play an important role in the transport of contaminants because of their reactivity and large surface area (Puls and Powell, 1992).

Collecting a representative sample of colloids is difficult because both drilling and pumping a well can create conditions that form colloids or mobilize them (McCarthy and Zachara, 1989; Ryan and Gschwend, 1990). Introduction of oxygen can change redox conditions; changes in temperature, pH, carbon dioxide partial pressure, and ion composition can alter the geochemical environment and cause colloid-sized precipitates to form during sampling (McCarthy and Zachara, 1989). Traditionally, concentrations of dissolved constituents have been determined by filtering water samples through 0.45-μm pore-size filters to separate the dissolved species from colloid-bound species. Because colloids can be smaller than 0.45 μm and, thus, may pass through the filters, concentrations of constituents in the dissolved phase can be overestimated.

Most water samples analyzed for mercury in southern New Jersey during the past 13 years were collected directly from a household tap after the water had run for 15 to 30 minutes; the water samples were not filtered. Thus, the mercury concentrations reported for most of the wells sampled are total concentrations, which could include both dissolved and colloidal mercury. Sampling of wells at one housing development in Egg Harbor Township, Atlantic County (site 2, reported in Barringer and others, 1997; see figure 1 for location) by the Atlantic County Division of Public Health and the NJDEP included the collection of several raw (unfiltered) and filtered ground-water samples at one home. The results of the study (table 1) indicate that from 36 to 54 percent of the mercury measured in the unfiltered samples was removed during filtration (John Dooley, New Jersey Geological Survey, written commun., 1993); whether the mercury remaining in the filtered samples was associated with colloids smaller than 0.45 μm (the pore size of the filters used) or was dissolved cannot be determined from these data.

In a previous study (Sites, 1994) of water from four wells at site 2 (including the well listed in table 1), the Atlantic County Division of Public

Table 1. Mercury concentrations in unfiltered and filtered ground-water samples from site 2, Egg Harbor Township, Atlantic County, New Jersey

[μg/L, micrograms per liter; site location shown in fig. 1]

Well identification number[1]	Date of sampling	Time of sampling	Total mercury concentration (μg/L)	Dissolved mercury concentration[2] (μg/L)
2062	06-13-91	0830-0850	17.8	8.2
2062	06-13-91	0950	18.6	9.9
2062	06-13-91	1135-1150	18.9	12.1
2062	06-13-91	1220	18.8	11.6

[1] Well identification number in Barringer and others (1997), which corresponds to identification number in unpublished New Jersey Department of Environmental Protection Ground Water Impact Area Report, June 1991, Trenton, N.J.

[2] Operationally defined; filtered through 0.45-micrometer membrane filter

Health and NJDEP collected samples that were passed through a variety of media in order to determine the most effective material for filtering out the mercury. The media included activated charcoal and several anion and cation resins; of these, all but a strong sodium cycle cation resin achieved a substantial decrease in total mercury concentrations. The results of this study indicate that the mercury present in elevated concentrations in ground water from the Kirkwood-Cohansey aquifer system may be present, at least in part, in a negatively charged form.

The data from well 2062 at site 2 (table 1), although limited, indicate that mercury adsorbed to colloids may represent a substantial portion of the mercury measured in the ground-water samples collected from that well. The effect that pumping in the residential neighborhoods may have had on local colloidal concentrations in ground water cannot be assessed easily, because wells at some sites have been sealed and homeowners are now receiving water from public-supply wells. When multiple water samples were drawn from several wells that yielded water with elevated mercury concentrations at various sites in southern New Jersey, the concentrations increased over time in 16 of 73 wells sampled three or more times. In others, concentrations remained relatively constant or decreased over time (fig. 2). A decrease in mercury concentration could be related to decreased domestic-well pumping in the neighborhoods after public-supply lines were extended to the developments; a decrease in the aggregate of pumped wells could result in fewer colloids being suspended in the water drawn from the aquifer. Both increases and decreases in mercury concentration could be related to the passage of a plume of contaminated water over time, or to a reconfiguration of the plume once pumping in the area lessened or ceased.

DESCRIPTION OF THE STUDY AREA

The study area lies within the northern Atlantic Coastal Plain, which extends from North Carolina to Long Island, N.Y. The Coastal Plain is underlain by a seaward-thickening wedge of predominantly unconsolidated sediments that extends eastward from the boundary of the Coastal Plain (the Fall Line) (fig. 3). The sediments range in age from Jurassic to Holocene (Trapp and Meisler, 1992). The Kirkwood-Cohansey aquifer system is

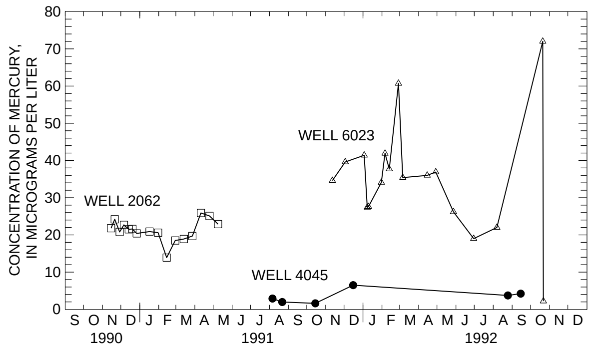
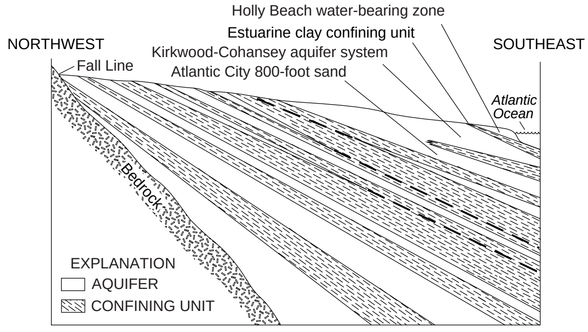


Figure 2. Variations in total mercury concentration with time in water from selected wells at sites 2, 4, and 6 in Atlantic County, southern New Jersey. (Site locations shown in fig. 1)



Modified from Martin, 1998

Figure 3. Generalized hydrogeologic section through the New Jersey Coastal Plain.

one of seven regional aquifers or aquifer systems recognized within the Coastal Plain; the sediments that compose it underlie about 3,000 mi^2 of southern New Jersey. Thus, it is the most areally extensive water-table aquifer in southern New Jersey, although it is confined on the Cape May peninsula (Zapecza, 1989).

Lithology

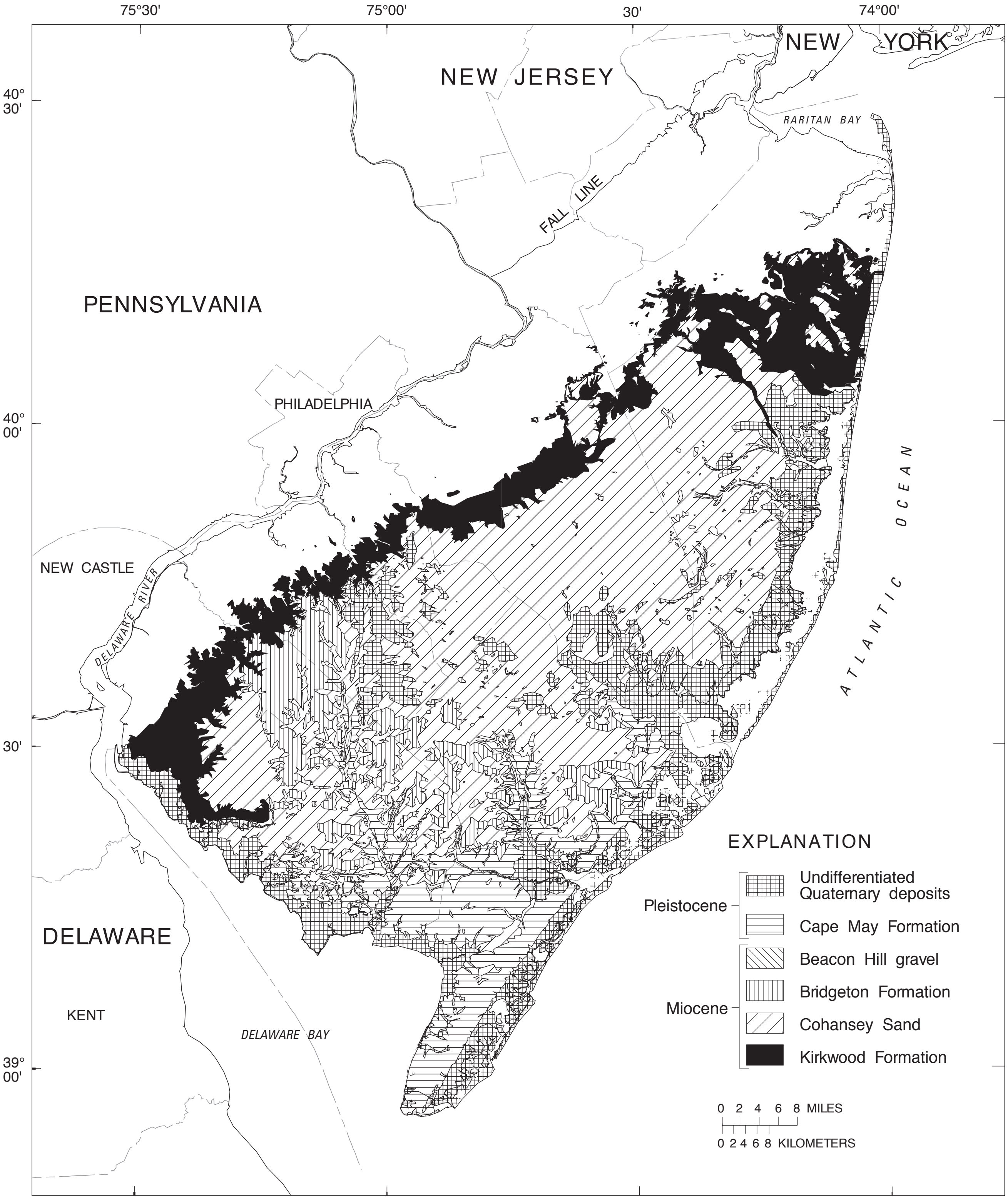
The two major lithologic units that compose the Kirkwood-Cohansey aquifer system are the Kirkwood Formation and the overlying Cohansey Sand, both of Miocene age. A discontinuous veneer of younger sediments overlies the Cohansey Sand; these younger sediments include the Beacon Hill Gravel and the Bridgeton Formation, of which the Bridgeton Formation is the more areally extensive (fig. 4). On the Cape May peninsula, the Pleistocene Cape May Formation overlies the Cohansey Sand (Zapecza, 1989).

The study reported herein is concerned only with the upper units of the aquifer system: the Cohansey Sand, and the overlying deposits, primarily the Bridgeton Formation. The Bridgeton Formation, which is the uppermost lithologic unit in many of the agricultural areas of southern New Jersey, is composed of highly weathered, coarse-grained arkosic sands and gravels that are composed primarily of limonite-stained quartz. Small amounts of weathered feldspar are present; the sediments are goethetic in some areas and contain nodules of aluminum oxide and ilmenite stringers. The Cohansey Sand is a quartz sand that grades from shallow marine to fluvial deltaic sequences across the coast (Owens and Minard, 1979). This limonite-stained sand also contains minor amounts of heavy minerals, particularly ilmenite and magnetite; clay minerals are primarily kaolinite and illite, and trace amounts of biotite mica are present.

Ground-Water Hydrology

The Kirkwood-Cohansey aquifer system is primarily an unconfined system; recharge to the aquifer system is from precipitation. The deep part of the system is recharged by precipitation principally in upland areas along the regional topographic divide that separates surface drainage to the Delaware River from drainage to the ocean, and discharges toward the outcrop area of the Kirkwood Formation to the west, to the Delaware Bay, and to the Atlantic Ocean.

Idealized local-scale hydrology in vertical section through central Burlington, Camden, Gloucester, and Salem Counties, where the sediments that make up the aquifer system are relatively thin, is shown in figure 5. Here, the Cohansey Sand is capped by the Bridgeton Formation in upland areas, and is hydraulically connected to it. The ground-water flow lines depicted diverge at the water-table divide and converge at the wetlands and stream. The uppermost flow line follows a much shorter path than does the lowermost flow line, which runs along the clay. Therefore, water



Base modified from U.S. Geological Survey digital
data, 1:100,000, 1983, Universal Transverse
Mercator projection, Zone 18

Geology from New Jersey Geological Survey
State Overlays

Figure 4. Geologic map of the sediments that crop out in the study area, New Jersey Coastal Plain.

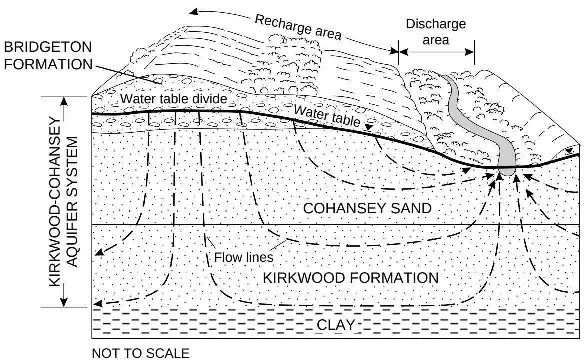


Figure 5. Idealized local-scale ground-water flow paths in central Burlington, Camden, Gloucester, and Salem Counties, southern New Jersey. (From Kozinski and others, 1995)

that enters at the divide takes longer to reach the discharge area than does water that enters farther downgradient and moves through the shallower part of the system. Furthermore, a contaminant that enters the aquifer near the divide will remain beneath its area of entry longer than will a contaminant that enters farther downgradient, where flow paths become horizontal at shallower depths. Locally, silty layers and clay lenses, which are common in the Cohansey Sand, alter flow paths, and may result in semiconfined or confined conditions over areas a few hundreds to a few thousands of feet in extent. Flow paths can be perturbed by pumping from wells; the perturbation is most pronounced when large-volume wells, such as public-supply wells, are withdrawing water.

Soils

The sediments of the Kirkwood Formation and the Cohansey Sand crop out over most of the seaward-sloping New Jersey Coastal Plain. The soils developed on these sediments typically are sandy, xeric, highly acid, and nutrient-deficient. The sand and silt fractions of these soils are dominated by quartz, which commonly is coated with hydrated iron oxides; A2 horizon soils typically are bleached. The soils typically contain small amounts of clay, and cation-exchange capacity is low (1-17 milliequivalents per gram) (Douglas and Trela, 1979). Uplands soils are well- to excessively well-drained, but soils developed on the Bridgeton Formation tend to contain weathered clay (kaolinite, aluminum-interlayered smectites and vermiculite), which increases their ability to retain moisture (Tedrow, 1979). Nevertheless, vegetable and fruit crops grown on soils that are farmed commonly are irrigated, and the strongly acid soils are limed (Markley, 1979).

With the exception of wetlands soils, whose organic-matter content is high, most surface soils in the study area contain relatively small amounts of organic matter (Markley, 1979, p. 84-85). Organic-matter concentrations in soils from 6 of the 34 sites of elevated concentrations of mercury in ground water ranged from none detected to 18.35 weight percent (Barringer and others, 1997). Soils from these sites were disturbed, with natural horizons obliterated or buried during the construction of the housing developments.

Land Use

Much of the study area falls within the approximately 1,700-mi^2 region referred to as the New Jersey Pine Barrens, or, more recently, the New Jersey Pinelands (New Jersey Pinelands Commission, 1980). Although large tracts of land currently are forested, the area’s natural resources have been used and the land surface altered, first by Native Americans, who set fires to drive deer herds and to clear the land, and then by European settlers from the late 17th century on. In the 18th century, farmland was located mostly in the area of the Cohansey River, which drains to the Delaware Bay. Interior areas were used for cattle grazing and lumbering; the native cedars were nearly exterminated by the demand for cedar shingles. Industries such as glass making and bog-iron mining and processing were scattered throughout the interior as well; these operations continued into the first half of the 19th century (Wacker, 1979). Many of the major towns and cities within the study area grew up around these early industries.

A shift in industrial activities occurred in the second half of the 19th century with the introduction of railroads as well as more economical methods of iron mining and processing elsewhere. Agricultural areas expanded and the population of the region grew (Wacker, 1979).

From 1900 to the World War II period, agriculture was a major industry in southern New Jersey. Areas along the Atlantic Coast became increasingly populated as railroads and the automobile made shore communities accessible for summer vacationers. Large areas of sparsely settled, reforested land persisted; several contained military bases, and some forested areas were set aside by the State as State Forests or various preserves. Residential areas expanded in southern New Jersey during the post-World War II period, commonly at the expense of farmland--a trend that has continued to the present.

Land-use history at the 34 sites of elevated mercury concentrations in ground water was determined by evaluation of aerial photographs dating to 1932 and USGS topographic quadrangles from the 1950's to the present (Barringer and others, 1997). About 75 percent of the residential developments were built on or adjacent to former or existing agricultural land.

Geochemistry of Water in the Kirkwood-Cohansey Aquifer System in Undeveloped and Agricultural Areas

Boxplots of selected constituents in water samples collected by the USGS from wells tapping the Kirkwood-Cohansey aquifer system in undeveloped and agricultural areas are shown in figure 6. Water from these wells also was analyzed for dissolved mercury, which was not detectable. The medians and ranges of calcium, magnesium, barium, strontium, and nitrate concentrations in water samples from the two land-use areas differ markedly. (Chemical and well-depth data for these wells are contained in appendix A.)

Undeveloped Areas

Water in the Kirkwood-Cohansey aquifer system in undeveloped areas is acidic, with pHs of typically about 5 or less, and with small, and sometimes negative, alkalinity (Barringer and others, 1993). Major cations such as calcium and magnesium typically are present in concentrations less than 2 mg/L, and sodium less than 3 mg/L. Potassium, an important plant nutrient, commonly is present at concentrations less than 1 mg/L. Concentrations of the major anions chloride and sulfate also are small, each being less than 5 mg/L (Barringer and Ulery, 1988), and nitrate, which is taken up by plants at the land surface, commonly is undetectable at the typical analytical limit of 0.01 mg/L (as nitrogen). Trace elements such as barium and strontium also are present at small concentrations, typically less than 100 and less than 20 µg/L, respectively. Overall, the chemistry of ground water that has not been affected by human activities is similar to that of precipitation, although dissolved constituents are slightly more concentrated. This is because the aquifer sediments and the soils developed on them contain relatively few weatherable minerals with which recharge and ground water can interact chemically.

The predominant mineral in the soils and the aquifer sediments is quartz. Iron hydroxide coatings on the sand and silt grains are relatively ubiquitous in the B horizons of Coastal Plain soils, and smectites and kaolinite also are present (Douglas and Trela, 1979). The quartz that forms the Cohansey Sand is goethitic; the overlying arkosic Bridgeton Formation sands and gravels are red and yellow from iron hydroxide coatings and may be cemented by iron hydroxides (Rhodehamel, 1979). Therefore, the potential exists for adsorption of trace elements, carried by recharge waters, to iron and aluminum hydroxides in the soils and aquifer sediments.

Carbonate minerals generally are lacking in soils and aquifer sediments of the New Jersey Coastal Plain, and clays are relatively sparse except where clay lenses occur; thus, few materials naturally present in the soils and sediments can neutralize the strong mineral acids in recharge that come from precipitation and the organic acids from vegetation such as pines. Consequently, the shallow ground water in undeveloped forested areas is nearly as acidic as the soils through which the recharge waters pass. Although dissolved organic carbon from fulvic and humic acids is present in elevated concentrations (greater than 2 mg/L) in many streams in the study area, particularly in bogs

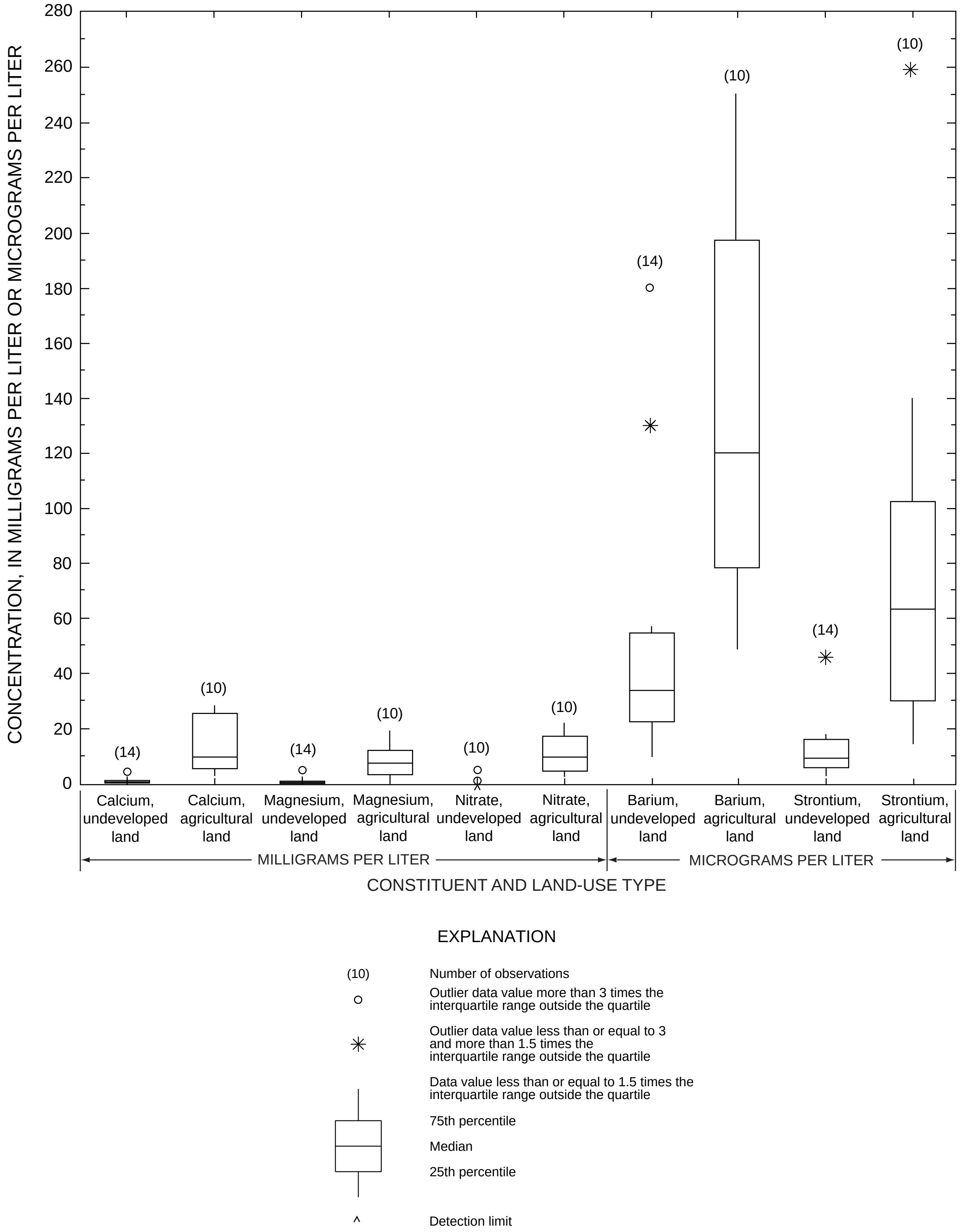


Figure 6. Distribution of calcium, magnesium, nitrate-nitrogen, barium, and strontium concentrations in water from the Kirkwood-Cohansey aquifer system underlying undeveloped and agricultural areas in southern New Jersey.

and fens (Lord and others, 1990), concentrations in ground water generally are less than 1 mg/L.

When land is developed, human activities commonly include the application of a variety of chemicals to the land surface or disposal of wastes on the surface and in the shallow subsurface. The chemistry of the water that passes through the unsaturated zone as recharge changes from that of the incident precipitation, and, ultimately, on a local scale, the chemistry of the ground water also changes.

Agricultural Areas

Agricultural land use probably provides the most clearly observed effect of land use on water quality because agriculture typically covers large areas, and substantial amounts of various chemicals can be applied to the land surface over many decades. In southern New Jersey, a commonly encountered effect is an increase in nitrate concentrations in ground water from agricultural areas over those observed in ground water associated with other land uses (Vowinkel, 1991). In southern New Jersey, applications of nitrogen fertilizer commonly exceed the ability of plants to utilize this nutrient. Consequently, nitrogen in the form of nitrate leaches from the soils into the aquifer. Nitrogen inputs from septic systems also can leach to ground water as nitrate (Miller, 1980). Unless the nitrate is reduced by bacterial or inorganic chemical reactions, it moves conservatively through the aquifer, transported as a solute by the ground water. Another conservative anionic constituent is chloride, which can be contributed by various agricultural chemicals and by road salt (Miller, 1980; Barringer and Ulery, 1988; Locat and Gelinas, 1989), and by septic systems (Miller, 1980). Additionally, calcium and magnesium concentrations typically are elevated in ground water underlying agricultural areas in southern New Jersey because the acidic soils are limed with dolomite to raise the pH. Barium and strontium are present in minor amounts in dolomite; the concentrations of these trace elements typically are elevated in ground water in agricultural areas as well (Kozinski and others, 1995).

Sulfate ion currently is a major constituent of precipitation, and probably has been so for much of this century, as it is supplied to the atmosphere by the burning of fossil fuels. Sulfate is also contributed to soils by some agricultural amendments, such as gypsum (Sposito, 1989). In an acidic geochemical environment, such as the sediments of Kirkwood-Cohansey aquifer system and the soils developed on them, sulfate may be adsorbed to clays and to iron hydroxides or may precipitate as sulfate salts, so that the ground water may not be enriched in sulfate.

STUDY METHODS

Field

Field activities during this study consisted of collecting aquifer sediments from a Camden County quarry for laboratory experiments, drilling boreholes at 2 of the 34 sites of elevated concentrations of mercury in ground water, using a screened auger to collect water samples from the borehole, and sampling selected wells at 3 of the 34 sites (locations of sampling sites shown in fig. 7). The water samples were analyzed for mercury and other chemical constituents; water samples also were collected at the two boreholes for age-dating by using two techniques, chlorofluorocarbon (CFC) dating and tritium/helium dating. The boreholes were installed so that water samples could be collected at various depths; results of analysis of the water samples were anticipated to show changes in concentrations of mercury and other constituents, water-quality characteristics, and age of water with depth. Water-quality samples at the three sites of elevated mercury concentrations were collected so that complete water chemistry at those sites could be assessed. Details of drilling, sample collection, and sample analysis are presented in appendix B.

Laboratory

A series of experiments was designed to evaluate the mobility of mercury in soils and aquifer sediments under a variety of chemical conditions. Adsorption experiments were performed to determine adsorption characteristics of dissolved

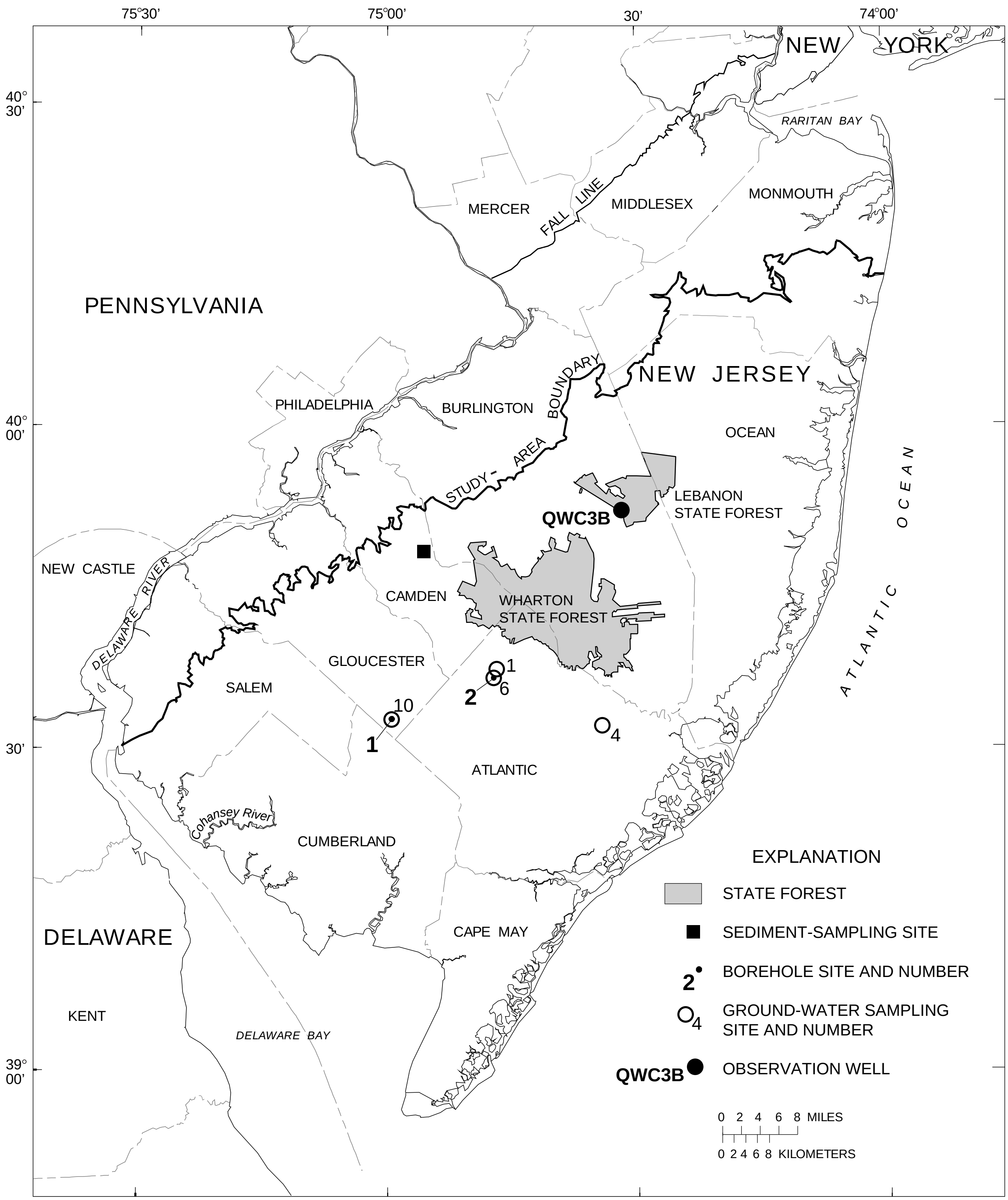


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Figure 7. Locations of sites for sediment-sample collection, borehole installation, and ground-water-sample collection, and location of observation well, Burlington, Camden, Gloucester, and Atlantic Counties, New Jersey.

mercury, mercuric chloride (Peterson, 1993), and PMA on sediments from the Cohansey Sand and the Bridgeton Formation. The sediment-size analysis, description of experimental procedures for the adsorption experiments, and raw adsorption data are in appendix C. Mass-balance calculations and a correction for pore fluids have been applied to the raw dissolved mercury and mercuric chloride adsorption data (Peterson, 1993) for the purposes of this report.

Additionally, preliminary desorption experiments were carried out by using columns in which mercury was desorbed from Cohansey Sand and Cohansey Sand overlain by Bridgeton Formation sediments. The leaching solutions included dilute (1×10^{-7} M and 1.4×10^{-4} M) nitric acid (HNO_3), two concentrations (1.4×10^{-2} M and 1.4×10^{-1} M) of sodium nitrate ($NaNO_3$), a nitrogen fertilizer solution (20-20-20), and a 0.1×10^{-1} M sodium chloride (NaCl) solution. A description of the apparatus and experimental procedures and the raw and calculated mercury-desorption data from the column experiments are presented in appendix D. Mercury analyses were performed in the Geology Department laboratory at Princeton University, Princeton, N.J., by using inductively coupled plasma spectroscopy (ICP) and cold vapor atomic absorption spectrometry (CVAAS) according to the method of Hatch and Ott (1968).

Adsorption Experiments

In order to determine the adsorption capacity of aquifer sediments for mercury, batch equilibrium experiments were performed with sediment samples from the Bridgeton Formation and Cohansey Sand. The solutions of mercury used in these experiments were composed of elemental mercury dissolved in 0.1-percent nitric acid and diluted with distilled water, mercuric chloride in distilled water (Peterson, 1993), and PMA in distilled water. The experiments were designed such that about 5 grams of sediment were placed in an acid-washed 50-mL polypropylene centrifuge tube and to this was added 25 mL of mercury solution at concentrations ranging from 10 to 100 µg/L. Experiments were carried out over a range of initial pHs (from 3.5-4.0 to 5.5-6.0); each set of experiments was performed in triplicate. Details of the experiments are presented in appendix C.

Previously, duplicates of adsorption experiments were conducted without sediments to check for mercury loss to or through vessel walls. Mercury loss was as much as 30 percent for the 10-µg/L solutions, but was less than 1 percent for the 100-µg/L solutions (Peterson, 1993).

Desorption Experiments

Cohansey Sand, with and without a veneer of Bridgeton Formation sediments, was used in a series of preliminary column experiments to assess the relative mobility of mercury adsorbed to the aquifer sediments when treated with sodium chloride, two concentrations of sodium nitrate, 20-20-20 fertilizer solution, and two concentrations of dilute nitric acid. Each leaching experiment was performed in triplicate. The columns were inoculated with a solution of mercury in 0.1-percent nitric acid diluted with distilled water to a concentration of 1,000 µg/L; the columns were drained after inoculation, covered, and allowed to equilibrate overnight (about 8-10 hours). The results indicate that mercury adsorption had reached a steady state in some columns (see app. D), but not in others. Following the inoculation, leaching solution was slowly added to the column. In both inoculant and leaching phases, the effluent and eluent, respectively, was collected (fig. 8), generally in 20-mL aliquots; these were analyzed to determine the mass of mercury that exited the column.

The mass of mercury in effluent from the inoculant phase was subtracted from the mass of mercury added to the column to determine the mass retained in the column. Because actual pore volumes varied from about 27 to 39 mL, it was not possible to correct accurately for inoculant solution remaining in the sediment pores by subtracting the first two aliquots (40 mL) of effluent collected. Without such a correction, both the amount of mercury adsorbed and the amount of mercury removed probably are slightly overestimated. Details and results of the experimental procedure are presented in appendix D.

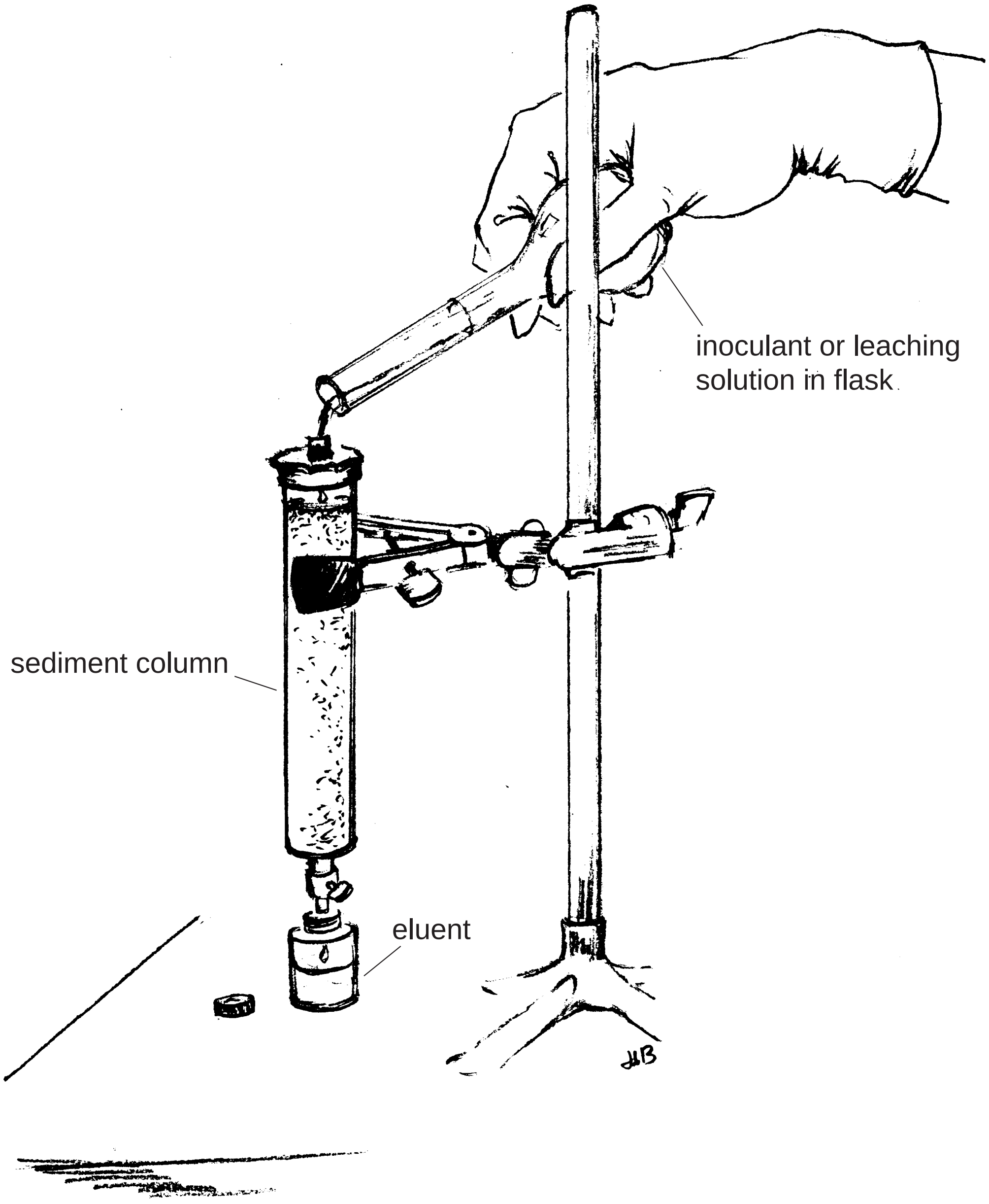

Figure 8. Diagram of column apparatus for mercury-desorption experiments.

Mercury was desorbed from the sediments by leaching with solutions of 1×10^{-2} M sodium chloride, 1 x 10^{-4} and 1.4×10^{-4} M nitric acid, 1.4×10^{-2} and 1.4×10^{-1} M sodium nitrate, and 20-20-20 fertilizer. Nitric acid is one of two important components of acid rain; the other is sulfuric acid. Only nitric acid was used in the preliminary experiments because large amounts of mercury are more soluble in nitric acid solutions than in water (most concentrated mercury standard solutions are made with nitric acid). The nitric acid solutions used were more acidic (pH 4.0 and 3.85) than the nitric acid component of acid rain reported by Dasch (1987), which is 3 x 10^{-5} M (pH 4.5). The pH of the more dilute nitric acid solution used in the experiments was consistent with the low pHs of acid rain measured in New Jersey (Lord and others, 1990).

Although the sulfuric acid component of acid rain may be important in mobilizing mercury, this was not tested. In these preliminary experiments, it was deemed more important to test interactions with the anions--nitrate and chloride--that are known to be present in elevated concentrations in ground water from areas where mercury concentrations also are elevated.

The fertilizer was obtained from a local agricultural supply and the solution prepared according to the directions on the label (about 3 g fertilizer in 1 L of water). Trace-element analysis was not provided; therefore, the solution was analyzed for mercury prior to beginning the experiments. The 20-20-20 fertilizer solution contained about 280 µg/L of mercury. To compensate for matrix effects, standards were mixed in a 20-20-20 fertilizer solution matrix. The manufacturer’s listed composition of the fertilizer and amount of mercury present are shown in table 2. Sodium chloride solutions were prepared with analytical-grade sodium chloride.

RELATION OF MERCURY TO OTHER CHEMICAL CONSTITUENTS

The relation of mercury to other chemical constituents could not be easily determined, as water samples from few wells had been analyzed for constituents other than mercury. Wells at 3 of the 34 sites were sampled during this study, but the distribution of mercury and other constituents with depth and, therefore, with age of water, could be determined only by sampling water from a range of

Table 2. Composition of 20-20-20 soluble fertilizer

[Average concentration of mercury in solution of fertilizer is 282 micrograms per liter; solution is about 0.4 oz fertilizer in 1 gal water (about 3 g/L)]

Component[1]	Percent	
Total nitrogen	20	
Ammoniacal nitrogen		3.90
Nitrate-nitrogen		6.15
Urea Nitrogen		9.95
Total phosphorous (Available phosphoric acid)	20	
Total potassium (Soluble potash)	20	

[1] Fertilizer components are derived from ammonium phosphate, potassium nitrate, and urea.

depths at a given location. Thus, the data collected from the two boreholes installed during the study were used to demonstrate how water quality changes with depth in the aquifer.

Vertical Distribution of Mercury and Other Constituents at Boreholes in Gloucester and Atlantic Counties

The results of analysis of water samples collected at seven different depths in borehole 1 in Gloucester County indicate that concentrations of dissolved mercury in ground water are elevated from about 60 ft to about 100 ft below land surface; that is, the mercury-contaminated water appears as a layer or lens in that depth interval. The results from borehole 2 in Atlantic County are less definitive because water could be sampled at only three depths. Mercury was detected in water drawn from 90 ft, but not in water from the two shallower depths.

Water samples from both borehole sites also were analyzed for major ions and other trace elements. The results of these analyses are described below. The concentrations of several constituents indicate that ground water has been affected by past human activities at the land surface.

Borehole 1--Gloucester County

Stiff diagrams of major ions in seven samples collected from 36 to 106 ft below land surface in borehole 1 on school property in Gloucester County are shown in figure 9. A diagram for water from a 36.6-ft-deep observation well in undeveloped land in Lebanon State Forest, Burlington County, is included for comparison. The major-ion chemistry of the water from borehole 1 at 36 ft shows little effect of human activities, although this water contains more dissolved constituents than ground water from undeveloped land as illustrated by the sample from the well in Lebanon State Forest. The Stiff diagrams show that concentrations of most of the major ions (calcium, magnesium, sodium, potassium, chloride, bicarbonate alkalinity) increased with depth and generally were greatest at 66 and 76 ft below land surface (see also table 3).

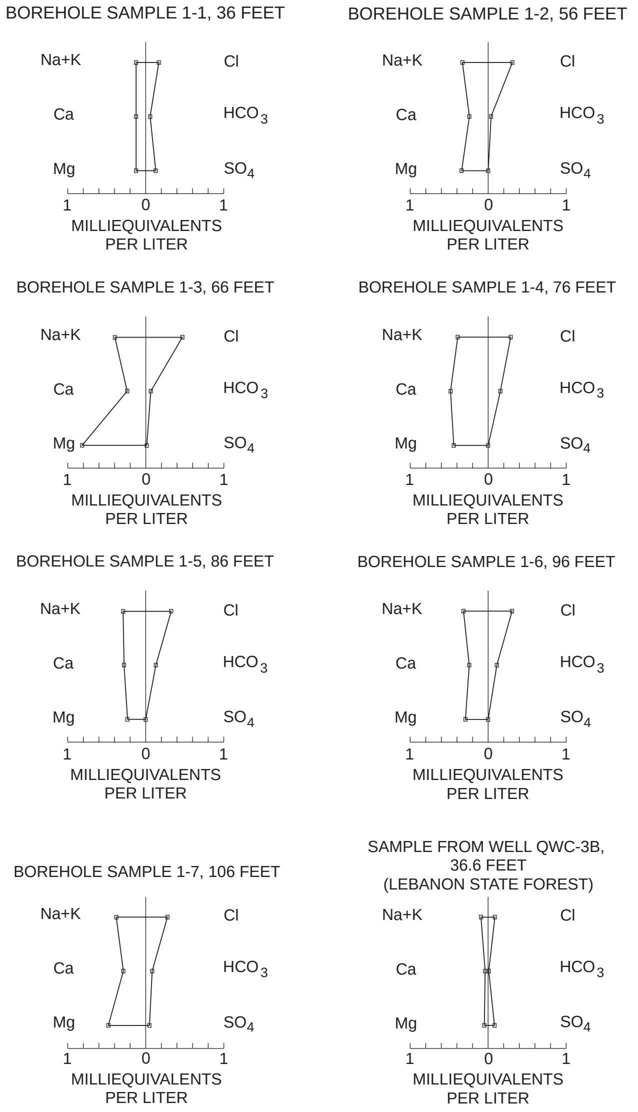

Figure 9. Stiff diagrams for seven water samples from borehole 1, Franklin Township, Gloucester County, and one water sample from a shallow observation well in Lebanon State Forest, Burlington County, New Jersey.

Nitrate concentrations also tended to increase with depth at borehole 1. Nitrate concentrations in water in the Kirkwood-Cohansey aquifer system in undeveloped areas typically are less than the reporting limit of 0.01 mg/L as nitrogen.

Table 3. Water-quality characteristics and constituent concentrations for ground-water samples from borehole 1, Franklin Township, Gloucester County, New Jersey

[ft, feet; µS/cm; microsiemens per centimeter at 25 degrees Celsius; mg/L, milligrams per liter; µg/L, micrograms per liter; $CaCO_3$, calcium carbonate; <, less than; --, not determined; location of borehole 1 shown in fig. 7]

Sample number	Depth below land surface (ft)	Turbidity (units)	pH (units)	Specific conductance (µS/cm)	Dissolved oxygen (mg/L)	Dissolved calcium (mg/L)	Dissolved magnesium (mg/L)	Dissolved sodium (mg/L)	Dissolved potassium (mg/L)
1-1	36	--	4.07	58	8.50	2.6	1.4	2.7	0.9
1-2	56	320	3.73	133	2.91	4.8	4.1	5.4	3.9
1-3	66	350	3.82	191	.56	4.8	10	6.7	4.5
1-4	76	52	4.21	177	.30	10	5.4	8.4	1.7
1-5	86	280	4.32	105	.37	5.5	2.8	5.7	1.2
1-6	96	26	4.23	116	4.90	4.9	3.6	5.7	2.8
1-7	106	17	4.56	165	5.28	5.7	5.9	5.8	4.8

Sample number	Dissolved chloride (mg/L)	Dissolved sulfate (mg/L)	Dissolved nitrate as N (mg/L)	Total alkalinity (mg/L as $CaCO_3$)	Dissolved iron (µg/L)	Dissolved manganese (µg/L)	Dissolved barium (µg/L)	Dissolved strontium (µg/L)	Dissolved mercury (µg/L)	Dissolved organic carbon (mg/L)
1-1	5.7	6.1	1.23	2.8	180	44	75	17	0.2	0.7
1-2	11	.2	8.63	2.2	260	140	310	48	<.1	2.1
1-3	16	.5	14.9	2.8	530	210	510	80	.2	2.2
1-4	9.9	.3	13.07	7.8	2,300	180	160	72	6.0	1.0
1-5	12	.2	4.88	6.9	2,700	49	74	34	28	.9
1-6	11	.2	6.48	5.5	1,300	27	120	43	6.0	.8
1-7	10	2.3	10.97	4.1	1,700	79	320	57	5.8	.6

Three of the seven samples from borehole 1 (66, 76, and 106 ft) contained nitrate concentrations in excess of the MCL (10 mg/L as nitrogen). Nitrate concentrations in samples of water from nearby wells in 1992 and earlier were below the MCL but greater than 6.0 mg/L; these concentrations are larger than expected background concentrations.

Land use at the site of experimental borehole 1, before school buildings were constructed there in the 1950's, was agricultural. The relatively small concentrations of nitrate and chloride (as well as major cations) in water at 36 ft below land surface indicate that recent input of these constituents has been negligible. Concentrations of calcium, magnesium, barium, and strontium measured in samples from 56 to 106 ft below land surface at borehole 1 were substantially larger than those in the sample from 36 ft, and also were substantially larger than median values reported in previous regional investigations of ground-water quality in the Kirkwood-Cohansey aquifer system (Barton and others, 1993; Harriman and Voronin, 1984). Bivariate plots of nitrate concentrations with concentrations of the divalent cations magnesium and barium indicate strong positive trends (fig. 10a and 10b); the trend of nitrate with calcium is less clear. Instead, the linear relation between nitrate and the monovalent cation sodium is strong (fig. 10c). Given the association of elevated concentrations of magnesium, barium, and strontium with elevated nitrate concentrations, the concentrations of these ions may be related to applications of agricultural chemicals, such as lime and fertilizer, in the past.

The presence of sodium in elevated concentrations, although consistent with agricultural inputs, also may be associated with septic-system inputs. Dissolved-oxygen concentrations were small between 66 and 86 ft below land surface (but not deeper); alkalinity was larger in the same depth interval than above or below, and nitrate concentrations also were large, although they decreased slightly with depth below this interval. The effect of the decrease in dissolved-oxygen concentrations, which could lead to nitrate reduction, may be offset by additional inputs of nitrate in water between 66 and 86 ft below land surface. The old septic system for the school was near the school buildings and could have been upgradient from

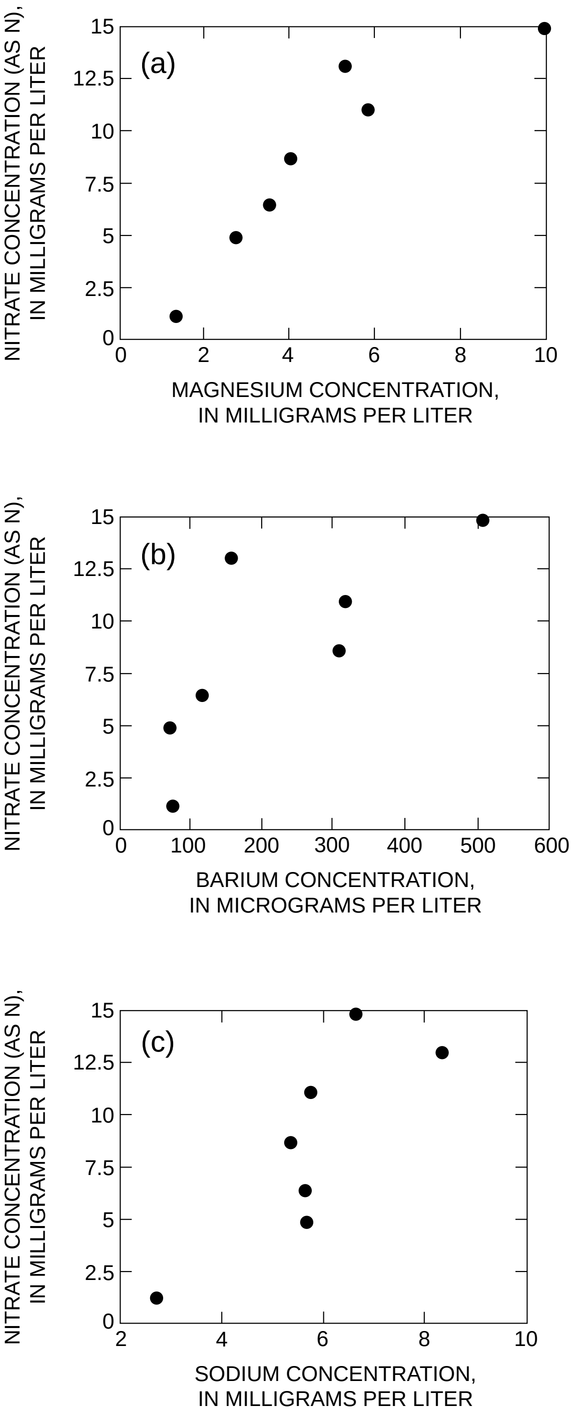

Figure 10. Variation in concentration of nitrate-nitrogen with concentrations of (a) magnesium, (b) barium, and (c) sodium in water samples from borehole 1, Franklin Township, Gloucester County, New Jersey.

borehole 1. (The present-day septic system is downgradient from school wells and the borehole.) The chemistry of water samples from borehole 1 may indicate agricultural inputs, reflected by constituent concentrations in samples from 96 to 106 ft below land surface, followed by septic-system inputs, reflected by constituent concentrations in samples from about 66 to 86 ft below land surface.

The distribution of mercury with depth at borehole 1 indicates that dissolved-mercury concentrations tend to be elevated in water containing elevated concentrations of calcium, magnesium, barium, nitrate and chloride, although relations between concentrations of dissolved mercury and those of most of the other dissolved constituents are not strong. The weak relations between mercury and many other dissolved constituents may be partly a result of the adsorption of some of the mercury to colloids that are removed during filtration, whereas constituents such as magnesium or chloride are not adsorbed to colloids and, therefore, are not removed by filtering. The role of colloids in the mobility of mercury has yet to be fully defined. Consequently, the coincidence of elevated mercury concentrations and elevated concentrations of conservative constituents such as chloride and nitrate could be the result of mercury-bearing colloids smaller than 0.45 µm moving at about the same velocity as dissolved constituents through the aquifer or of the presence of mercury as a mobile complex.

Ranked dissolved-mercury concentrations show a strong positive correlation (correlation significant at the 0.05 level) with ranked dissolved-iron concentrations (fig. 11). This relation can be interpreted in two ways: (1) mercury is adsorbed to iron hydroxide colloids small enough to pass through the 0.45-µm filter, or (2) mercury initially adsorbed to iron hydroxides may be released as iron is reduced and goes into solution. Given that dissolved-oxygen concentrations decreased substantially between 56 and 86 ft below land surface, where dissolved-iron concentrations were largest, this latter interpretation is possible.

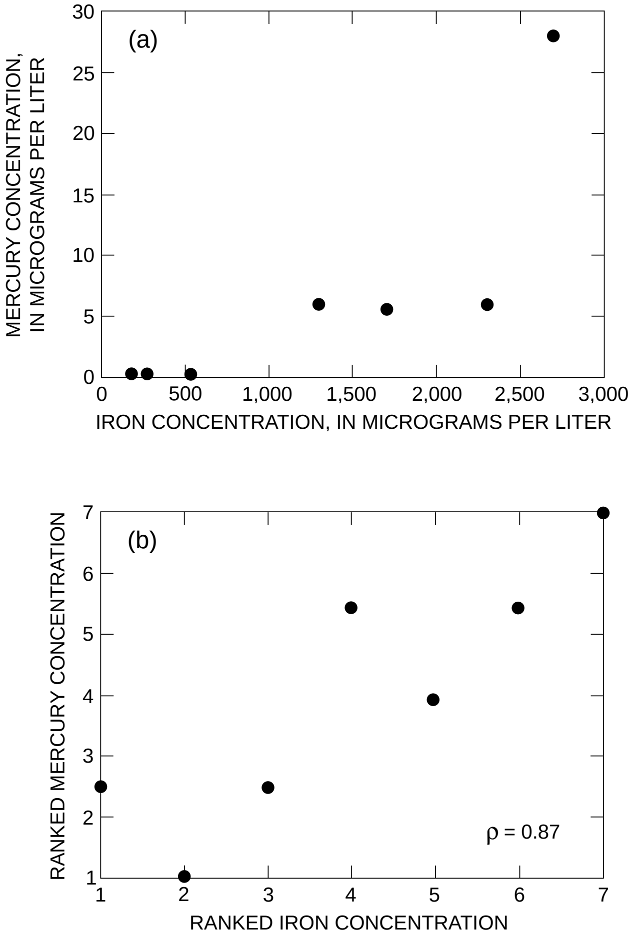

Figure 11. (a) Raw and (b) ranked concentrations of mercury and iron in filtered water samples from borehole 1, Franklin Township, Gloucester County, New Jersey. (The Spearman's rho correlation coefficient for the ranked data is 0.87, which is significant at the 0.05 level.)

The distribution of dissolved mercury and redox-sensitive species with depth (fig. 12) indicates that redox conditions may affect mercury mobility. Dissolved-oxygen concentrations decreased with depth to 86 ft, whereas iron concentrations increased and manganese concentrations increased and then decreased. Manganese concentrations increased at shallower depths than iron concentrations, indicating that reduction of manganese in hydroxide coatings on aquifer sediments may occur at slightly higher dissolved-oxygen concentrations than reduction of iron in hydroxide coatings. This is consistent with the redox chemistry of Mn and Fe; the manganous ion is stable at more oxidizing conditions than the ferrous ion

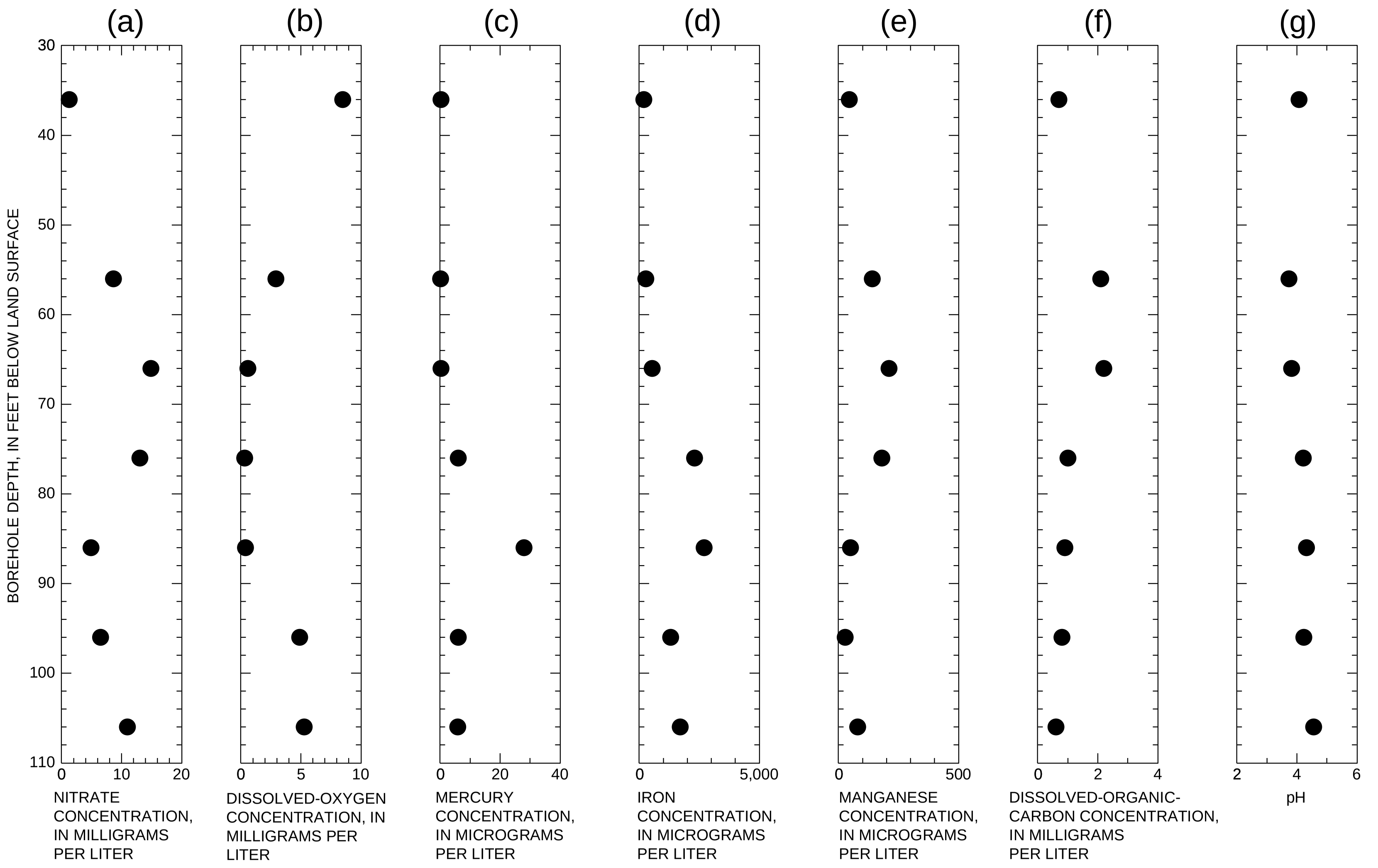

Figure 12. Concentrations of (a) nitrate-nitrogen, (b) dissolved oxygen, (c) dissolved mercury, (d) dissolved iron, (e) dissolved manganese, and (f) dissolved organic carbon with depth, and (g) pH with depth in water samples from borehole 1, Franklin Township, Gloucester County, New Jersey.

(Krauskopf, 1967). The coincident increases in pH and dissolved-iron concentration also are indicative of iron reduction and dissolution, because the reaction consumes hydrogen ion. On the basis of these data, mercury sorbed to iron hydroxide coatings appears to be released to solution as iron is reduced and goes into solution.

The concentrations of nitrate observed in water from borehole 1 are a function of nitrate inputs from the surface or subsurface as well as redox reactions; therefore, increases and decreases in nitrate concentrations in the ground water may not be related solely to the changes in redox conditions that are indicated by the concentrations of constituents such as iron and manganese that are, presumably, naturally occurring. Whether the changing redox conditions indicated by constituent concentrations in water from borehole 1 are part of the geochemical environment in the aquifer at the other sites of elevated concentrations of mercury in ground water is unknown.

Borehole 2--Atlantic County

Borehole 2 was installed at a site where 10 wells had yielded water with mercury concentrations exceeding the MCL; one well, upon repeated sampling, had yielded water with concentrations as large as 72 µg/L, although these levels subsequently decreased. Drilling was more difficult at the site of borehole 2 than at the site of borehole 1 because many silty layers were encountered. Water samples were obtained from 30 and 60 ft below land surface, but deeper drilling was precluded by the presence of clay and silt in the aquifer sediments. An adjacent monitoring well screened 90 ft below land surface was sampled to obtain geochemical information from greater depths in the aquifer. Results of analyses of these water samples are shown in table 4.

Mercury was not detected in the two shallower samples, and was present in only a small concentration (0.2 µg/L) in the deepest sample. Virtually no well-construction data are available for the domestic wells yielding mercury-contaminated water at this site, with the exception that one well is known to be 90 ft deep. Wells in the vicinity draw water from 64 to 105 ft below land surface (Ground Water Impact Area Report, 1993, on file at New Jersey Department of Environmental Protection, Trenton, New Jersey); therefore, it is possible that the mercury-contaminated water is tapped mostly by wells deeper than 90 ft, or that the contaminated water has moved over time in response either to pumping or to natural ground-water flow within the aquifer. In any case, the monitoring well did not appear to tap water containing elevated mercury concentrations on September 23, 1993. On April 6, 1993, however, water from this well contained a total-mercury concentration of 5.50 µg/L. Because the later sample was filtered and the earlier sample was not, most of the mercury may have been colloidal and, therefore, retained on the filter in the September 1993 sampling.

The samples from 30 and 60 ft below land surface were turbid compared with the sample drawn from the monitoring well and iron concentrations, although measured in a filtered sample, were largest in the two most turbid samples. Dissolved-iron concentrations were largest in samples with very small (0.63 mg/L) and moderate (3.52 mg/L) dissolved-oxygen concentrations, and were smallest in the sample with a small (0.89 mg/L) dissolved-oxygen concentration. Thus, the effects of redox conditions and (or) colloids on iron concentration are not easily discernible. Iron-hydroxide colloids may be a factor in the concentrations of metals measured in these samples. Water from the 30- and 60-ft depths did not contain detectable dissolved mercury; as in the case of the 90-ft monitoring well, colloids containing both iron and mercury may have been removed by filtering.

The major-ion chemistry of ground water at borehole 2 is similar to that at borehole 1. At borehole 2, however, water from the shallowest interval sampled (30 ft below land surface) showed clear evidence of the effects of human activities: nitrate, sodium, and chloride concentrations were elevated and the nitrate concentration exceeded the MCL. The elevated sodium and chloride concentrations may reflect inputs of road salt to the aquifer--borehole 2 was installed in a residential area, about 15 ft from one road and about 200 ft from another, intersecting road. Nevertheless, in addition to the elevated nitrate concentration, the dis-

Table 4. Water-quality characteristics and constituent concentrations for ground-water samples from borehole 2, Hammonton Town, Atlantic County, New Jersey

[ft, feet; µS/cm; microsiemens per centimeter at 25 degrees Celsius; mg/L, milligrams per liter; µg/L, micrograms per liter; $CaCO_3$, calcium carbonate; <, less than; location of borehole 2 shown in fig. 7]

Sample number	Depth below land surface (ft)	Turbidity (units)	pH (units)	Specific conductance (µS/cm)	Dissolved oxygen (mg/L)	Dissolved calcium (mg/L)	Dissolved magnesium (mg/L)	Dissolved sodium (mg/L)	Dissolved potassium (mg/L)
2-1	30	410	4.49	234	0.63	0.8	1.7	34	3.9
2-2	60	240	4.71	132	3.52	4.6	5.6	6.8	1.8
2-3	90	1.4	4.56	131	.89	5.4	4.7	7.4	3.2

Sample number	Dissolved chloride (mg/L)	Dissolved sulfate (mg/L)	Dissolved nitrate as N (mg/L)	Total alkalinity (mg/L as $CaCO_3$)	Dissolved iron (µg/L)	Dissolved manganese (µg/L)	Dissolved barium (µg/L)	Dissolved strontium (µg/L)	Dissolved mercury (µg/L)	Dissolved organic carbon (mg/L)
2-1	20	5.0	13.01	9.6	2,200	52	110	11	<0.1	1.6
2-2	12	1.3	7.16	9.8	1,700	39	79	59	<.1	.8
2-3	9.7	.3	10.41	5.0	83	57	200	67	.2	.4

solved-oxygen concentration was small and the dissolved-iron concentration was large in water 30 ft below land surface (about 10 ft below the water table). Although the borehole is about 200 ft from a stream and wetlands, the reducing conditions close to the water table are indicative of septic-system inputs to shallow ground water. The deeper water sample (from 60 ft below land surface) was substantially more oxygenated than the shallower sample; the deepest sample (from the 90-ft well) contained substantially less dissolved oxygen, a condition that is not uncommon in deep ground water. The vertical distribution of dissolved oxygen indicates the presence of zones of differing redox conditions; the upper low-oxygen zone may be related to septic-system inputs and the lower low-oxygen zone may be the result of loss of oxygen through chemical reactions as ground water moves farther from the unsaturated zone.

Both undeveloped and agricultural land were present at the site of borehole 2 before residential development began in the 1960’s. The elevated concentrations of major ions in water at various depths at borehole 2 could be derived from two different sources, the chemistry of deeper water reflecting inputs of agricultural chemicals from earlier agricultural land use, and the chemistry of shallower water reflecting septic-system inputs from the later residential development.

Age of the Water Sampled at the Boreholes

Water samples were collected from boreholes 1 and 2 for CFC and tritium/helium ($^3H/^3He$) age dating. The results of the CFC analyses could not be used to determine the age of the water because, although previous indications were that ground water at the borehole sites did not contain VOCs, VOC contamination of the samples collected from the boreholes was apparent. The compounds detected in water from the two boreholes were different; therefore, it is unlikely that the samples were contaminated as a result of inadequate steam cleaning of the auger between sites. Because the presence of VOCs in the water samples precluded an accurate determination of the CFC peaks on the chromatogram, these data are not reported.

Ages were determined by tritium/helium age dating for 7of 10 water samples; results are given in table 5. These values are preliminary in that they have not been corrected for temperature and for the helium/neon ratio. These corrections could change the ages by ± 2 or 3 years (Stefan Drenkard, Lamont-Doherty Geological Observatory, oral commun., 1995).

The ages determined for water samples from borehole 1 are, for the most part, consistent with those estimated using by a vertical flow velocity of 1 m/yr for the Kirkwood-Cohansey aquifer system (Zoltan Szabo, U.S. Geological Survey, oral commun., 1994), although the deeper samples were slightly younger than expected. The ages determined for water samples from borehole 2 and the adjacent monitoring well are about half those estimated from assumed flow velocities, and half

Table 5. Tritium/helium ages of ground water at different depths in borehole 1, site 10, Franklin Township, Gloucester County, and borehole 2, site 6, Hammonton Town, Atlantic County, New Jersey

[ft, feet; ^3H, tritium; ^3He, helium; T.U., tritium units*; -, data not available]

Sample number	Depth below land surface (ft)	^3H (T.U.)	$^3H + ^3He$ (T.U.)	Estimated age (years)
1-1T	33-36	-	-	-
1-2T	53-56	-	-	-
1-3T	63-66	14.7 ± 0.5	40	17.5 ± 2
1-4T	73-76	24.4 ± .7	100	25.5 ± 2
1-5T	83-86	17.6 ± .5	85	28.5 ± 2
1-6T	93-96	17.1 ± .5	92	30.5 ± 2
1-7T	103-106	-	-	-
2-1T	28-30	22.4 ± .7	25	2.5 ± 2
2-2T	58-60	14.2 ± .4	23	9.5 ± 2
2-3T	80-90	18.36 ± .1	-	15 ± 1.5

*A tritium unit is defined as an abundance of one atom of tritium per 10^{18} atoms of hydrogen; it is equivalent to 7.1 disintegrations of tritium per minute per liter of water (Faure, 1986).

those of water in borehole 1 at similar depths. Depth to water was about the same (about 17 ft) at boreholes 1 and 2, but, because the sediments at borehole 2 generally were more silty than those at borehole 1, water in borehole 2 was expected to be older than water at similar depths in borehole 1. The discrepancy in ages of ground water between the two sites may be a consequence of difference in pumping at the sites. The school well near borehole 1 has a larger capacity than any of the individual domestic wells at borehole 2. Nevertheless, more than 50 domestic wells in the vicinity of borehole 2 were being pumped or had recently been pumped when the samples were collected. The aggregate effect of the pumped wells may have been to draw younger water more deeply into the aquifer at borehole 2 than did withdrawals from the school well at borehole 1.

The ages of water determined for samples from borehole 1 can be used to estimate a date when mercury-contaminated water would have first entered the aquifer as recharge. Borehole 1 at site 10 is near the local topographic divide, which is presumed to coincide with the local ground-water divide; therefore, ground-water flow paths probably are nearly vertical in the area of the borehole. Depth to water was 17 ft in September, when the water table generally is low. A conservative average annual depth to water probably is about 15 ft. Mercury-contaminated water at site 10 was found as deep as 120 ft from the land surface in one of the school wells there, but it is not known whether deeper water is contaminated as well. Therefore, the earliest date to which mercury movement in recharge from the surface can be linked is about 1945. This is determined in the following manner. First, it is assumed that recharge moves rapidly (on the order of a few days) through the unsaturated zone to the water table. The thickness of the saturated zone at the 120-ft well would be about 105 ft. On the basis of ages determined for water from depths of 66 to 96 ft below land surface, a difference of 13 years in ground-water age spans a vertical distance of 30 ft. Consequently, the average annual rate of vertical movement is about 2.3 ft. Subtracting 45.7 years (105 ft / 2.3 ft/yr) from 1991, the year when the water in the 120-ft well was first sampled and mercury detected, gives 1945 as the approximate date when mercury-contaminated recharge entered the system.

This estimate is subject to several constraints: first, that the age/distance relation is constant with depth; second, that the mercury moves at the same rate as the ground water; and third, that mercury contamination had just reached 120 ft below land surface in 1991. The 1945 estimated date is applicable only to mercury in water at 120 ft; in fact, because there are no mercury data for water from wells in the area deeper than 120 ft, it is not known to what depths the mercury-contaminated water extends. Nor is the date of the contaminant's arrival at 120 ft known. Therefore, it is possible that the onset of mercury movement from the land surface can be linked to a date earlier than 1945. Because the elevated mercury concentrations are present at the same depths as elevated chloride and nitrate concentrations, mercury appears to have moved at the same rate as these conservative constituents. Without sampling to determine whether this geochemical relation is true for ground water at depths greater than 120 ft, the apparent movement of mercury together with other constituents related to human activities cannot be confirmed.

Relations Between Concentrations of Mercury and Other Constituents at Selected Sites

In the first phase of this investigation (Barringer and others, 1997), 34 sites were identified where one or more wells had yielded water containing total mercury concentrations greater than 1 μg/L, a concentration that is two orders of magnitude greater than typical background concentrations measured by Windom and Smith (1992). Of the 34 sites, all but 2 included one or more wells that yielded water with mercury concentrations above the MCL. In many cases, the water samples collected by State and county agencies also had been analyzed for VOCs; several organic compounds, including benzene, toluene, trichloroethylene (TCE), tetrachloroethylene (PCE), and chloroform, were detected in some of the samples. Of these, chloroform was nearly ubiquitous, but no pattern of occurrence of the other compounds was apparent. Other chemical characteristics and constituents, such as pH, major ions, or trace elements other than mercury, rarely were measured in these water samples.

Subsequently, water samples were collected during this investigation at selected wells at three of the sites of elevated mercury concentrations in ground water (see fig. 7) and were analyzed for dissolved major ions, nutrients, and trace elements, as well as pH, specific conductance, and dissolved oxygen to generate a suite of water-quality data that could be used to identify chemical anomalies that are associated with particular sources of chemical inputs to ground water. The analytical results are shown in table 6.

The water-quality data for the three sites show fundamental differences between concentrations of constituents at site 1 and concentrations at sites 4 and 10 (fig. 13). Some characteristics of water samples from site 1, such as small specific-conductance values and small calcium, magnesium, potassium, barium, and strontium concentrations, are more similar to those in water from undeveloped areas than to those in water underlying agricultural areas. Nitrate concentrations in water samples from three wells at site 1 were slightly larger (1.1-3.2 mg/L) than would be expected for ground water from undeveloped areas. Furthermore, the two wells that yielded water with elevated mercury concentrations of 1.3 and 4.5 mg/L are the same wells that yielded water with the largest chloride concentrations measured at site 1, in addition to the slightly elevated nitrate concentrations. Water from well 1084, which contained the largest mercury concentration, contained the largest concentrations of calcium, magnesium, potassium, barium, and strontium, constituents typically associated with agricultural land use. Water from the other well at site 1 in which mercury concentrations were elevated (well 1117) also contained the largest concentrations of sodium and chloride of the five wells sampled (18 and 23 mg/L respectively). These concentrations are substantially larger than those found in ground water from undeveloped areas, and are large even for ground water underlying some residential areas. Such concentrations may be indicative of road salt inputs to the aquifer, but sodium and chloride input from septic-system effluent cannot be discounted.

Water from all of the wells sampled during this study at site 4, and most of the wells sampled at site 10, exhibits water-quality characteristics that typically are associated with agricultural effects on ground water. At site 4, in particular, a strong positive relation between mercury concentrations and concentrations of magnesium, barium, and nitrate was found (fig. 14). Past land use at site 4 was predominantly agricultural with some residential areas, a mosaic of land uses that has changed little over the past 5 or 6 decades.

Three wells at site 10 (10003, 10019, and 10022) yielded water that shows little effect from agriculture or other human activities. Well 10003 is a shallow (29 ft), recently drilled monitoring well at the school. Results of analysis of water samples from borehole 1 at the same site, reported earlier, indicate that shallow, recently recharged ground water at this site does not show the effects of agriculture or other human activities. The other two wells are near older residences that appear to be downgradient from the school, where the largest concentrations of mercury in well water have been measured. The house associated with well 10022 was the site of a small thermometer factory; the concentration of mercury in the soil beneath the exhaust fan at the back of the house is large (258 µg/kg) relative to concentrations of mercury measured in area soils during this study. The mercury measured in the soil beneath the exhaust fan presumably is related to past manufacturing activities; however, no mercury has been detected in water from either the domestic well or a shallow monitoring well at the house.

Positive relations of magnesium concentrations to chloride, nitrate, and barium concentrations are strong in water from the wells at site 10, and calcium is strongly positively related to potassium (fig. 15). Like those in water from borehole 1, dissolved-mercury concentrations in water from other wells at site 10 are not strongly related to concentrations of any of the above constituents, but concentrations of mercury are elevated in water in which these other constituents also are elevated. The limited data on the spatial distribution of mer-

Table 6. Water-quality characteristics and constituents for water samples from wells at three sites of elevated mercury concentrations in ground water in Atlantic and Gloucester Counties, New Jersey

[ft, feet; µS/cm; microsiemens per centimeter at 25 degrees Celsius; mg/L, milligrams per liter; µg/L, micrograms per liter; $CaCO_3$, calcium carbonate; <, less than; --, no data available]

Well-identification number[1]	Depth below land surface (ft)	pH (units)	Specific conductance (µS/cm)	Dissolved oxygen (mg/L)	Dissolved calcium (mg/L)	Dissolved magnesium (mg/L)	Dissolved sodium (mg/L)	Dissolved potassium (mg/L)	Dissolved ammonia as N (mg/L)
Site 1 (Atlantic County)									
1050	85	4.0	43	4.2	0.21	0.43	2.8	0.45	0.01
1083	--	4.7	35	5.5	.39	.48	2.1	.42	.01
1084	104	4.4	74	6.3	1.1	1.2	6.0	.58	<.01
1114	--	4.5	35	7.7	.58	.65	2.2	.30	<.01
1117	--	4.4	109	7.5	<.02	<.01	18	.17	.01
Site 4 (Atlantic County)									
4001	110	4.4	112	0.3	2.4	2.8	9.9	1.41	<.01
4047	--	4.4	113	.8	2.2	2.5	8.0	1.34	<.01
4048	--	4.4	92	3.4	1.3	1.7	8.8	1.07	.01
4050	105	4.5	166	.3	2.3	2.4	21	1.37	<.01
Site 10 (Gloucester County)									
10001	120	4.5	125	4.9	4.3	4.5	7.3	1.9	<.01
10002	90	4.8	113	3.4	4.3	3.5	6.0	1.5	<.01
10003	29	4.7	146	7.5	5.2	3.2	9.3	.90	<.01
10004	60	4.8	121	7.5	3.4	3.5	6.8	1.2	<.01
10012	60	4.6	146	8.0	4.2	5.8	7.8	1.4	<.01
10013	--	4.4	196	8.5	7.4	10	4.5	2.0	<.01
10014	--	4.7	271	7.1	7.4	10.7	12	3.6	<.01
10019	--	4.5	84	--	.6	1.9	4.3	.71	<.01
10022	--	5.0	37	7.6	<.5	.5	1.8	.25	<.01
10023	26	4.6	230	--	14.2	1.7	5.5	5.6	.12
10029	--	4.7	176	4.5	6.5	6.9	8.0	4.0	<.01
10030	86	5.3	107	.6	3.4	3.1	6.6	--	--
10031	--	4.5	181	9.2	7.5	8.4	6.5	1.9	<.01

Table 6. Water-quality characteristics and constituents for water samples from wells at three sites of elevated mercury concentrations in ground water in Atlantic and Gloucester Counties, New Jersey--Continued

Well-identification number[1]	Dissolved chloride (mg/L)	Dissolved sulfate (mg/L)	Dissolved nitrate as N (mg/L)	Total alkalinity (mg/L as $CaCO_3$)	Dissolved iron (µg/L)	Dissolved manganese (µg/L)	Dissolved barium (µg/L)	Dissolved strontium (µg/L)	Dissolved mercury[2] (µg/L)	Dissolved silica (mg/L)
Site 1 (Atlantic County)										
1050	5.3	0.3	.43	--	37	3	21	5	0.1	4.9
1083	3.9	.4	.22	4.1	92	5	17	5	.1	4.4
1084	7.4	.3	3.2	2.7	7	16	45	15	4.5	5.8
1114	2.9	.3	1.1	3.8	<3	12	25	5	.2	5.3
1117	23	.3	1.1	2.2	<3	<1	<2	<.5	1.3	4.1
Site 4 (Atlantic County)										
4001	13	0.3	5.8	4.4	60	20	160	40	12.5	7.3
4047	10	.3	5.3	3.5	29	15	150	35	11.3	7.5
4048	14	.3	2.3	3.7	4	35	110	20	3.4	7.2
4050	30	5.9	2.9	3.8	38	16	130	35	3.7	8.6
Site 10 (Gloucester County)										
10001	--	--	--	--	11	27	160	53	19.4	8.3
10002	8.7	<.1	6.8	5.1	149	36	142	40	20.6	4.3
10003	20	13	1.8	3.3	21	50	148	20	<.2	2.9
10004	9.4	<.1	8.0	2.3	86	71	258	35	<.2	2.6
10012	15	<.1	9.0	2.2	74	101	363	47	2.5	3.1
10013	26	<.1	11	1.3	40	74	448	76	<.2	4.0
10014	27	<.1	16	2.1	66	126	501	91	.4	3.0
10019	8.8	<.1	3.9	3.4	57	16	90	9	<.2	2.2
10022	4.3	<.1	.3	3.1	47	4	29	4	<.2	2.3
10023	9.2	27	4.4	1.4	65	118	47	65	<.2	4.4
10029	21	<.1	11	2.0	68	49	331	60	.2	3.0
10030	--	--	--	--	3,000	48	55	37	2.0	7.9
10031	20	<.1	12	1.3	20	47	264	76	.3	3.1

[1] All wells are domestic wells except 10001, 10002, 10030, which are school wells, and 10003, 10023, which are shallow monitoring wells. Well numbers correspond to numbers in Barringer and others (1997).

[2] Raw mercury-concentration data, measured at the Princeton University Geology Department Laboratory, are presented in Barringer and others (1997, app. 2g and 2h).

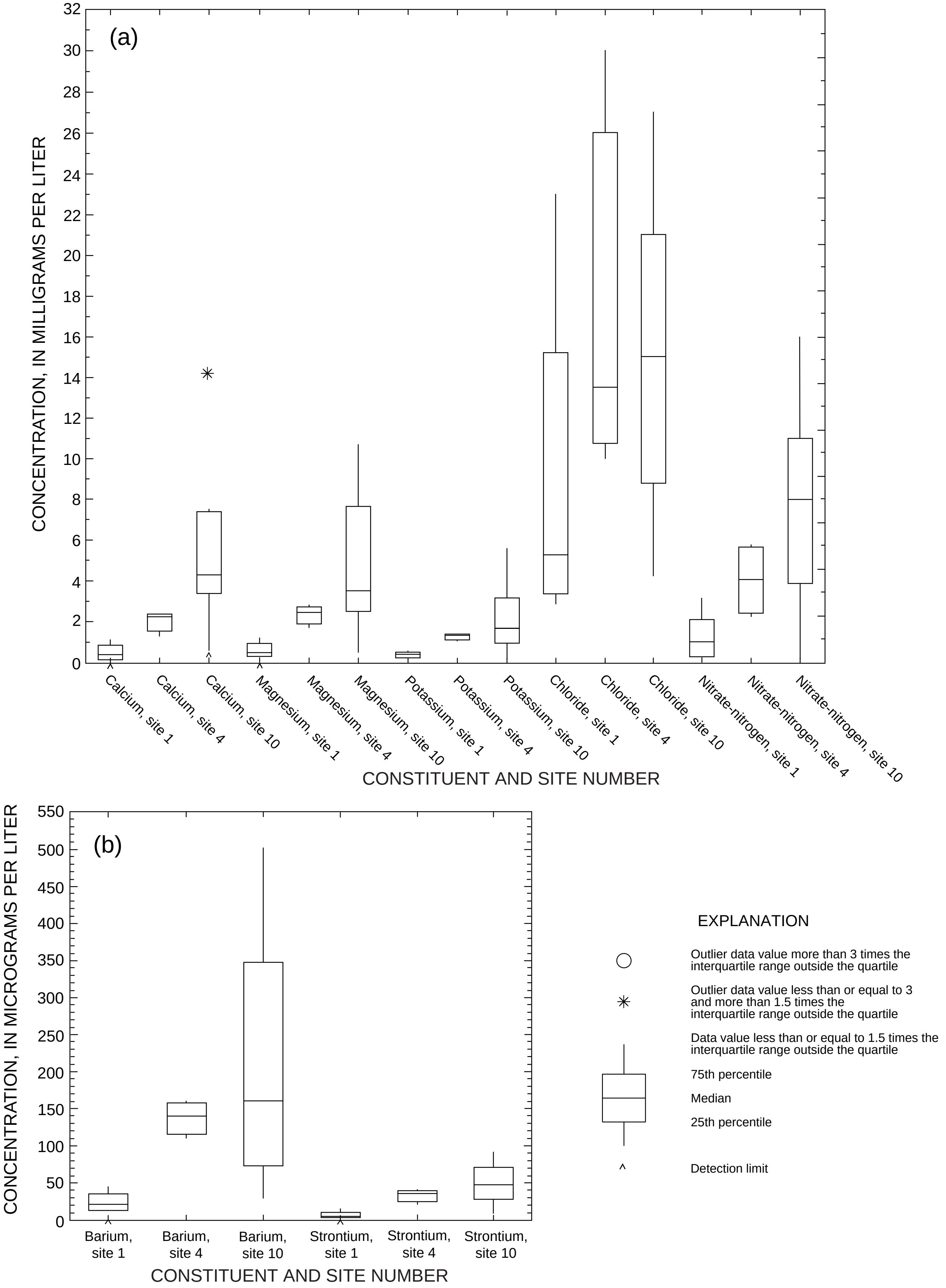

Figure 13. Distribution of concentrations of (a) calcium, magnesium, potassium, chloride, and nitrate-nitrogen and (b) barium and strontium in ground water from sites 1, 4, and 10, Atlantic and Gloucester Counties, New Jersey. (Site locations shown in fig. 1)

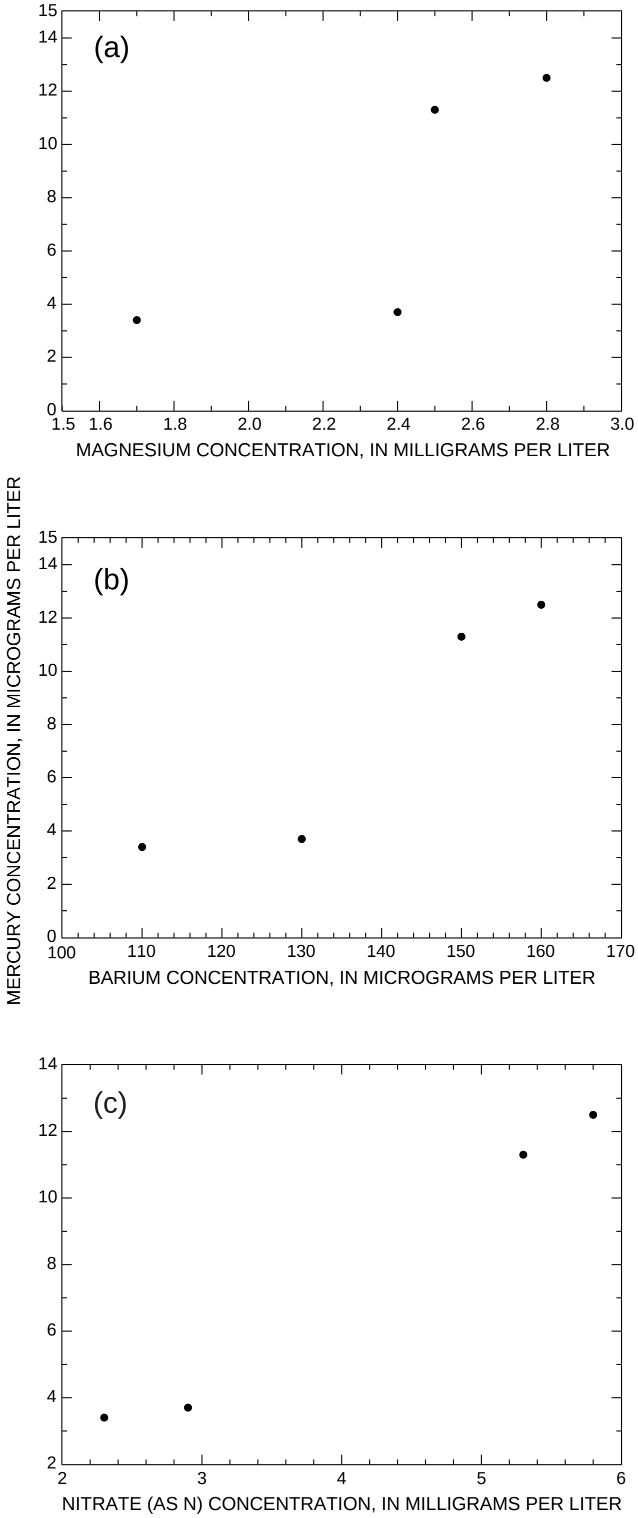

Figure 14. Relation between mercury concentrations and concentrations of (a) magnesium, (b) barium, and (c) nitrate-nitrogen in ground water from site 4, Atlantic County, New Jersey. (Site location shown in fig. 1)

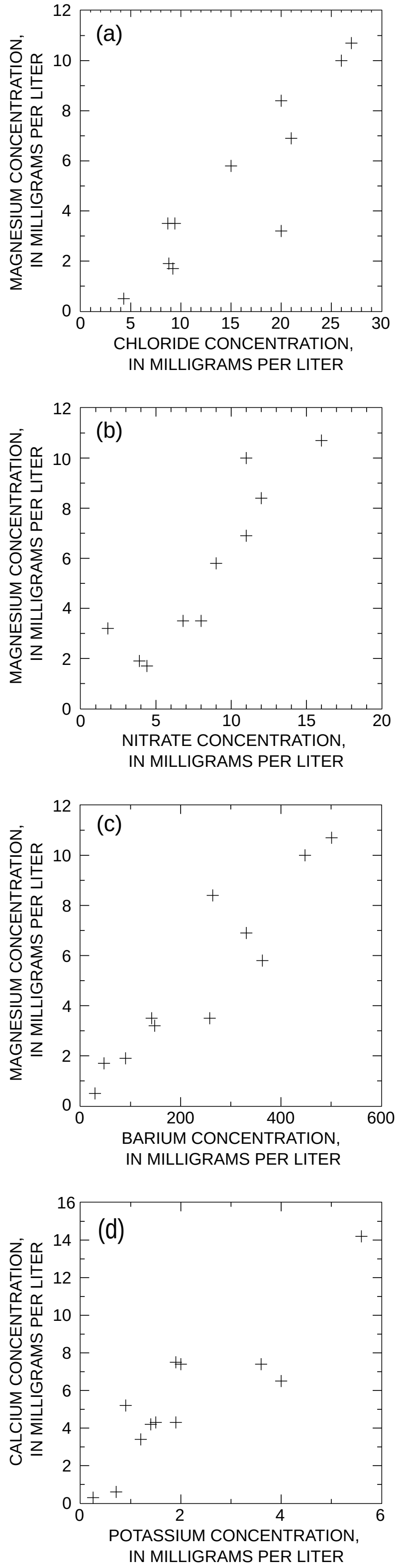

Figure 15. Relations between magnesium concentrations and (a) chloride concentrations, (b) nitrate-nitrogen concentrations, and (c) barium concentrations, and (d) relations between calcium concentrations and potassium concentrations in water from wells at site 10, Gloucester County, New Jersey. (Site location shown in fig. 1)

cury at site 10 indicate that concentrations in water from the school wells are significantly larger than elsewhere at site 10; therefore, the source of the mercury probably was near the school. The ground water at sites 4 and 10 appears to have been affected by agricultural activities; this assessment is corroborated by information on past land use gathered from aerial photographs, topographic maps, and discussions with residents. Nevertheless, because some of the same constituents supplied by agricultural chemicals are present in septic-system effluent, chemical effects from this latter source also are possible.

The chemistry of ground water at site 1 is more difficult to explain in terms of past land use. Although earlier aerial photographs of site 1 and the vicinity were not available, photographs taken in 1962 show that the housing development was being built and the surrounding land was partly forested and partly farmland. Site 1 is located on the shore of a lake that probably receives ground-water discharge. Wells at the houses may tap ground water that originated in an upland area and is now nearly at the end of a series of regional flow paths. It is also possible that the density of pumped wells in this development has been sufficiently great that water flows from the lake into the aquifer near the lake shore.

Analytical results for water samples from the five wells sampled at site 1 during this study indicate that concentrations of constituents associated with agriculture (calcium, magnesium, barium, strontium, and nitrate) were small in water from four of the five wells sampled and were, for the most part, in the range expected for water in undeveloped areas (see fig. 6). Although the nitrate concentrations were above the generally undetectable background levels typical of ground water from undeveloped areas, they were smaller than many of the concentrations commonly found in water underlying agricultural areas. After the mercury contamination was found at site 1, NJDEP sampled 38 wells at site 1 for selected constituents other than mercury; the nitrate concentrations measured generally were similar to those measured during this study, with concentrations typically less than 5 mg/L (R.A. Gallagher, New Jersey Department of Environmental Protection, written commun., 1995). Only 3 of the 38 wells sampled by NJDEP yielded water with concentrations of nitrate greater than 5 mg/L. Chloride concentrations were greater than 10 mg/L in water from these three wells (background concentrations typically are 3 to 5 mg/L), but such elevated concentrations are not unique to these three wells. Ten of the 38 wells at site 1 yielded water with chloride concentrations greater than 10 mg/L, indicating that road salt could be a factor in mobilizing mercury at site 1. Because no sodium-concentration data are available for the 38 wells, this hypothesis cannot be confirmed. Again, elevated chloride concentrations also may be derived from septic-system effluent. Analysis of ground-water samples for detergents would be useful in determining whether septic-system effluent could be a source of the elevated chloride concentrations in ground water from site 1, or whether road salt is the likely source.

No known point source for the mercury at site 1 has been found; the only possible point source, identified through aerial photograph interpretation, is the site of a World War I munitions factory about 2 miles from site 1. Local hydrology, as shown by a previously developed water- table map (Clark and Paulachok, 1989), indicates that ground-water flow from the site of the former munitions factory probably is not toward site 1. A detailed flow-path analysis of site 1 would be needed to “backtrack” mercury along flow paths from current locations of elevated mercury concentrations to discover the source.

Because water-quality data from other sites of elevated mercury concentrations in ground water generally are sparse, associating chemical characteristics of ground water with elevated mercury concentrations was more difficult elsewhere than at sites 4 and 10. Site 6 (see figs. 1 and 7) is a housing development about 1 mi south of site 1 in Atlantic County, and is the location of borehole 2. The water-quality data from the borehole sampling indicate chemical effects that may be related to inputs of agricultural chemicals, septic-system effluent, road salt, or some combination of these sources, as discussed earlier. Other available water-quality data tend to confirm these findings. Results of analyses of samples from 27 domestic wells for nitrate (R.A. Gallagher, written commun., 1995;

Randi DiMartini, Atlantic County Division of Public Health, written commun., 1993) and 21 domestic wells for chloride (R.A. Gallagher, written commun., 1995) indicate that nitrate concentrations generally were greater than background concentrations, and in 12 samples were in the range from 3 to 22 mg/L (table 7). Most of the chloride concentrations were larger than background concentrations; 16 of the 21 wells yielded water with chloride concentrations ranging from 5 to 25 mg/L. Water levels in monitoring wells at site 6 indicate that septic-system input may be causing local mounding of the water table (R.A. Gallagher, written commun., 1995). Water-table mounding could be occurring at other sites where the density of pumped wells and of septic systems is high. Local mounding of the water table resulting from septic-system recharge in addition to slight local depressions in the water table around each pumped well could increase the hydraulic gradient from septic systems toward some wells. A conceptual model of this phenomenon is shown in figure 16. The hydraulics of this effect currently are being examined through computer simulations with a finite-difference ground-water flow model (Shilpa Rajagopalan, University of Pennsylvania, oral commun., 1999).

At site 16 (see fig. 1), a formerly agricultural residential neighborhood in Atlantic County, concentrations of mercury in water from 13 percent of the domestic wells sampled exceeded the MCL. Most of the water samples also were analyzed for VOCs, but few were detected. A subset of water samples from site 16 also was analyzed for nitrate; of 29 samples, nitrate concentrations in 12 met or exceeded the MCL (10 mg/L), and concentrations in 13 others were greater than the typical background concentration of less than 0.01 mg/L (table 8). The largest mercury concentrations did not coincide with the largest nitrate concentrations.

At site 25 (see fig. 1), a residential neighborhood in Cumberland County, concentrations of mercury in water from 20 percent of the domestic wells sampled exceeded the MCL. Additional water-quality data collected from eight wells at and near site 25 (table 9) show that nitrate concentrations also are elevated in water from these wells and exceed the MCL in water from three of them. Barium concentrations also are larger than those in water underlying undeveloped land, and are similar to concentrations in water underlying agricultural land (fig. 6). Ranked concentrations of nitrate are positively correlated with ranked barium concentrations (correlation significant at the 0.05 level). Mercury concentrations are not normally distributed and, as at site 16, do not correlate strongly with nitrate concentrations. Although the area at and near site 25 was partly residential in 1953, forested areas and open areas with long structures, possibly barns, and orchards were present at the site during the 1950's, the 1960's, and the early 1970's. Residential development increased during those decades and into the 1980's. The nitrate and barium concentrations could be indicative of agricultural effects on the ground water; additional chemical data for the area surrounding and including this site would be needed to accept or reject this hypothesis.

At those sites where agricultural effects on ground-water quality are indicated by the water chemistry (sites 4, 6, 10, and 25), the pH of the ground water is low, typically less than 5. Acidic ground water is present even at those sites where elevated calcium, magnesium, barium and strontium concentrations provide possible evidence for liming of soils. Although soil pHs may be raised by additions of "lime" (dolomite), the low pH of the underlying ground water appears to be maintained by the release of hydrogen ion during nitrification of ammonia in fertilizers (Kozinski and others, 1995). Ammonia is the dominant form of nitrogen in septic-system effluent (Miller, 1980, p. 218); nitrification causes acidic conditions in septic-system plumes as well (Robertson and others, 1991). The effect of pH on dissolved mercury complexes has been discussed earlier, and adsorption experiments, discussed below, indicate that pH plays a role in the adsorption of various forms of mercury.

Table 7. Water-quality data for water samples from 27 wells at site 6, Atlantic County, New Jersey, 1991-93

[µg/L, micrograms per liter; mg/L, milligrams per liter; <, less than; --, no data; all samples collected by New Jersey Department of Environmental Protection unless otherwise indicated; well numbers correspond to those in Barringer and others, 1997)]

Well-identification number	pH	Mercury (µg/L)	Nitrate (as N) (mg/L)	Chloride (mg/L)	Dissolved solids (mg/L)	Hardness (mg/L)
6001	5.08	2.6	2.82	10.5	63	12
6003	5.72	<.2	.18	1.0	54	12
6004	5.34	<.2	1.89	4.5	38	12
6008	5.26	6.7	1.99	3.5	44	12
6013	5.19	<.2	5.25	9.0	80	30
6017	5.6	3.0	1.10	10.0	<25	4
6019	5.1	4.49	1.16	5.0	32	11
6021	6.9	13.4	3.0	10.0	122	--
6022	4.6	4.19	6.2	25.0	87	4
6023	5.0	34.6	3.9	11.0	25	19
6027	6.0	2.64	3.9	6.0	<25	10
6029*	--	.33	22.2	--	--	--
6033	6.0	2.91	4.44	17.0	77	10
6034	5.09	<.2	4.51	10.5	85	16
6035	6.15	4.3	3.14	3.5	71	24
6038*	--	<.2	16.6	--	--	--
6045	5.0	6.96	2.5	9.0	37	9
6046	5.07	<.2	1.2	5.0	46	16
6051	5.39	<.2	3.15	8.0	84	22
6054	5.2	.81	1.46	6.0	35	11
6060*	--	<.1	1.41	--	--	--
6061	5.42	.2	0.31	1.5	27	12
6062	4.86	6.1	1.78	17.5	48	20
6064	4.92	<.2	6.24	7.0	83	32
6069*	--	.3	<0.1	--	--	--
6071*	--	.1	0.29	--	--	--
6072*	--	.5	0.71	--	--	--

*Water sample from this well collected and analyzed by Atlantic County Division of Public Health

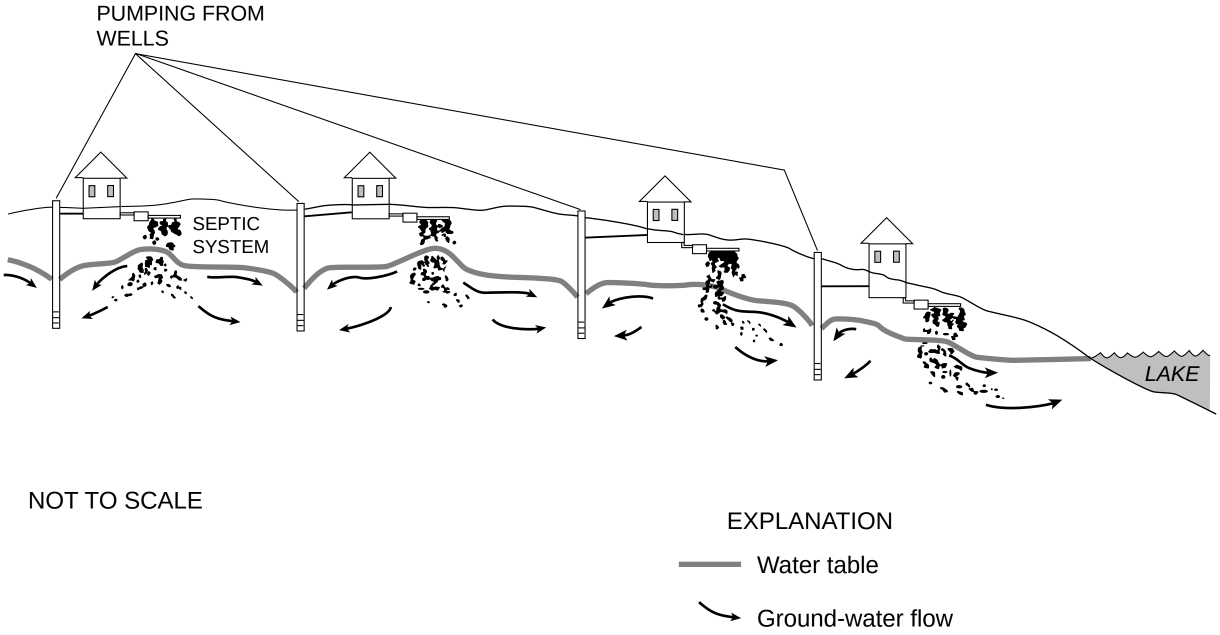

Figure 16. Conceptual model of water-table mounding beneath septic systems and water-table depression around wells.

Table 8. Mercury and nitrate concentrations in water samples from 29 wells at site 16, Buena Vista Township, Atlantic County, New Jersey, 1983-92

[µg/L, micrograms per liter; mg/L, milligrams per liter; <, less than; data from Atlantic County Division of Public Health and New Jersey Department of Health Environmental Chemistry laboratory]

Well-identification number[1]	Total nitrate as N (mg/L)	Total mercury (µg/L)	Well-identification number[1]	Total nitrate as N (mg/L)	Total mercury (µg/L)
16001	9.00	1.04	16050	1.40	0.22
16002	14.6	4.16	16051	3.10	1.48
16003	14.8	2.70	16052	2.70	.22
16004	18.8	.31	16053	.10	.43
16005	1.40	.71	16054	7.20	.12
16006	10.0	1.37	16055	3.43	2.99
16007	<.10	.57	16056	<.10	<.10
16008	4.98	.48	16059	2.50	3.68
16009	1.85	.94	16060	17.5	.18
16010	24.4	.67	16063	6.40	5.73
16011	10.6	.85	16066	5.80	.30
16012	14.7	1.06	16069	7.60	1.09
16013	12.5	<.10	16075	15.9	1.00
16014	11.8	.65	16076	18.2	2.90
16015	<.10	.26			

[1] Well numbers correspond to those in Barringer and others (1997)

Table 9. Water-quality data for water samples from eight wells at and near site 25, Vineland City, Cumberland County, New Jersey, 1987

[mg/L; milligrams per liter; µg/L, micrograms per liter; <, less than; all data from City of Vineland Health Department]

Well-identification number*	pH (units)	Total nitrate as N (mg/L)	Total barium (µg/L)	Total mercury (µg/L)
25035	4.62	15	280	4.7
25034	4.32	9.8	80	<.5
25028*	4.67	14	70	<.5
25033	4.93	5.6	50	<.5
25001*	4.48	9.8	90	17
25032	4.45	10	110	2.3
25031	4.31	9.0	70	<.5
25036	4.21	5.4	87	14

*Well number corresponds to that in Barringer and others (1997)

Table 10. Grain-size measurements as a percent of total sediment mass for sediments of the Cohansey Sand and the Bridgeton Formation, Camden County, New Jersey

[mm, millimeters; >, greater than; <, less than; data from Peterson (1993)]

Sediment size fraction	Grain size (mm)	Sieve size (U.S. mesh)	Cohansey Sand (percent)	Bridgeton Formation (percent)
Granule	>2.00	10	0.12	0.02
Very coarse sand	1.00	18	2.2	.61
Coarse sand	.50	40	61.58	29.98
Medium sand	.25	60	27.37	51.19
Fine sand	.125	120	6.69	17.66
Very fine sand	.074	200	.59	.22
Coarse silt	.034	400	.70	.13
Silt/clay	<.034	pan	.75	.19

MOBILIZATION OF MERCURY FROM LAND SURFACE OR SUBSURFACE TO GROUND WATER

Mercury applied at the land surface first must pass through soils and sediments to reach the water table. In order to reach the depths within the aquifer at which elevated concentrations of mercury have been measured, it must remain mobile as it passes through unsaturated aquifer sediments to the water table, or be mobilized repeatedly. In order to understand how mercury introduced into the study-area soils or aquifer sediments can be mobilized, the adsorption capacities of the soils and sediments needed to be evaluated so that basic questions regarding transport mechanisms could be addressed. Perhaps the most fundamental question is--given the sandy soils and aquifer sediments in the study area, does mercury deposited on the land surface move rapidly through those soils and sediments in the unsaturated zone and into ground water, or does it adsorb to the soils and sediments? If it does adsorb, it would need to be mobilized by change in the physical and (or) geochemical environment, induced by disturbance and (or) the presence of some added chemical agent, in order to reach the aquifer.

Adsorption to and Desorption from Soils

Adsorption of mercury to samples from 15 New Jersey soil series was studied by Yin and others (1996). Of these, three soil series--Downer, Lakewood, and Sassafras--commonly are present in the study area. All three soil series are acidic (Markley, 1979); the pH of the samples collected by Yin and others (1996) ranged from 4.18 for the Lakewood sand to 5.78 for the Sassafras sandy loam.

Adsorption experiments using a 1×10^{-7} M solution of mercury were carried out at pH values ranging from 3 to 10. Mercury (II) was adsorbed extensively to the soils studied at pHs from 3 to 5 (Yin and others, 1996). Because the samples were ground to pass a 2-µm sieve (Yin and others, 1996, p. 838), surface coatings on sand grains undoubtedly were disturbed and fresh grain faces exposed. Thus, adsorption characteristics of the individual soil series probably are not quantitatively comparable, but can be used to derive general principles regarding adsorption of mercury to the study-area soils. The results of Yin and others (1996) generally are similar to those discussed by Barrow and Cox (1992), which show that adsorption of hydrated mercury ion is favored at pHs near 4. A more recent study (Yin and others, 1997) of adsorption kinetics of mercury on A-horizon soils indicates that organic carbon enhances adsorption.

Kinetics of mercury desorption from A-horizons of four soil series, one of which is common in the Coastal Plain, were studied by Yin and others (1997). Mercury was adsorbed from two solutions (4 and 8 mg/L) until equilibrium was reached. Desorption was initiated by applying a 0.01 M $NaNO_3$ solution to the soils. The soils and solution in the reaction chamber were stirred continuously in later experiments, whereas earlier experiments were done in batch mode. Desorption of mercury was greater when the solution was stirred than in the batch experiments. (This observation could indicate that soil or sediment disturbance may affect mercury mobility.) Yin and others (1997) also found that organic carbon inhibited desorption of mercury.

Adsorption to and Desorption from Aquifer Sediments

Characteristics of Aquifer Sediments

The Cohansey Sand and Bridgeton Formation sediments are primarily medium- to coarse-grained sand (table 10), which is predominantly quartz and which commonly is coated with iron hydroxides. Of the sediments used in the adsorption (Peterson, 1993; this study) and desorption (this study) experiments, those of the Cohansey Sand are slightly coarser than those of the Bridgeton Formation. Differences between the two sedimentary units are more apparent in the fine-grained fractions. The Cohansey Sand sediment samples contained more silt and clay than did the Bridgeton

Formation samples (1.45 and 0.32 percent of the total sediment mass, respectively). The content of organic carbon as measured by loss on ignition was low for both sediments--0.04 mass percent for Cohansey Sand sediments and 0.70 mass percent for Bridgeton Formation sediments (Peterson, 1993). If these values are representative, the aquifer sediments contain much less organic matter than many of the study-area soils, which contained from 0.37 to 18.35 mass percent (Barringer and others, 1997).

The silt/clay-sized fraction of the Cohansey Sand contained significant amounts of kaolinite with lesser amounts of illite, and minor amounts of quartz and feldspar. Iron hydroxides, some amorphous and some in the form of goethite, were a major component of the Bridgeton Formation silt/clay-sized fraction. Analysis of the fine fractions by X-ray diffraction also indicated the presence of minor amounts of plagioclase feldspar and degraded kaolinite (Peterson, 1993).

Adsorption to Aquifer Sediments

Adsorption data from a series of batch equilibrium experiments were evaluated to determine the distribution coefficients for mercury, mercuric chloride, and PMA on sediments of the Cohansey Sand and Bridgeton Formation. These are the parent sediments from which the overlying soils have developed; they comprise the lower part of the unsaturated zone as well as all (or part, in the case of the Bridgeton Formation sediments) of the saturated part of the aquifer considered in this report.

Because it was assumed that mercury in various forms could be mobilized from New Jersey Coastal Plain soils, three forms of mercury were selected for adsorption experiments: elemental mercury (Hg^o), mercuric chloride ($HgCl_2$), and PMA ($C_8H_8HgO_2$). Elemental mercury is a decomposition product of organomercurial compounds used as agricultural and lawn pesticides and as mildew inhibitors in paint; elemental mercury also is discharged as waste by some laboratories, hospitals, and dental offices, and by some industrial activities. Mercuric chloride was used on row crops and as a seed dressing during the first half of the 20th century. PMA has been used as a fungicide on fruit trees and turf, as an herbicide to kill crabgrass, and as a seed dressing.

Of the three mercurial forms or compounds used, mercuric chloride is the most soluble in water and elemental mercury the least. Although PMA can begin to decompose to elemental mercury within a few minutes, Kimura and Miller (1964) found that 60 to 70 percent of the PMA applied to an agricultural soil was still present in that form after 28 days. Thus, it is possible that all three initial forms of mercury used in the experiments could leach through soils and reach unsaturated aquifer sediments.

The form of the mercury present at the conclusion of each batch experiment was not determined; therefore, it is not known if any conversion or decomposition of the adsorbent took place during the experiment. It is assumed that the elemental mercury, which was dissolved in 0.1 M nitric acid and then diluted, was oxidized by the acid. How much of the mercury in those diluted solutions was Hg^0 (aq) and how much was a species containing Hg (II) was not calculated. Therefore, the solutions formed by dissolving elemental mercury are referred to in this report as “dissolved-mercury solutions.” All solutions used in the batch experiments were chosen to span an order of magnitude in mercury concentrations. Mercury concentrations chosen were 10, 20, 50, 70 or 80, and 100 µg/L (Peterson, 1993; this study).

The results of the adsorption experiments were plotted as S as a function of C, log S as a function of log C, and 1/S as a function of 1/C (where S = the concentration adsorbed (µg/kg) and C = the concentration in solution (µg/L)) in order to determine whether a Langmuir or Freundlich model best fit the data. The Freundlich adsorption isotherm ($S = KC^n$, where $n = 1$) is the same as the linear isotherm ($S = K_dC$), and incorporates the assumption of an infinite amount of solute that can be adsorbed (Fetter, 1993). Where $n \neq 1$, the data can be linearized by plotting log S against log C. It is unwise to extrapolate beyond a linear relation shown by the data because the relation may not continue to be linear. In the use of the Langmuir isotherm model, however, a finite number of adsorption sites is assumed; the formulation of the two-surface Langmuir model can accommodate data that plot as two straight-line segments (Fetter, 1993).

In a test of equilibrium and kinetic models, Amacher and others (1986) used both a two-surface Langmuir and a Freundlich isotherm model to describe the adsorption of mercuric chloride on different soils. Hogg and others (1978) used a Langmuir model to estimate the maximum adsorption capacity of several forms of mercury on two soils. In the present study, it was found that the Freundlich model best described the experimental results; therefore, the Freundlich equation ($n = 1$) was used to calculate the distribution coefficients (K_ds) for the mercury compounds on the sediments (MacLeod and Peterson, 1993). All mass balances in this report were calculated by using raw data from Peterson (1993); furthermore, results of all adsorption experiments (including those with PMA) were corrected for fluid remaining in the sediment pores. Therefore, the distribution coefficients reported here differ from previously published distribution coefficients.

Results of the batch experiments indicate that between about 38 and 95 percent of the mercury put into the experimental system (by using solutions containing 10 to 100 μg/L of mercury) adsorbed to the sediment. The amount of mercury adsorbed is calculated on the basis of the assumption that the concentration of mercury in the liquid not captured during centrifugation is the same as that in the liquid captured. The volume captured is corrected for the volume of liquid remaining in the pores after centrifugation; therefore, the original volume of liquid is used in the calculation of the amount of mercury adsorbed. The adsorption of the various forms of mercury to the aquifer sediments is illustrated in figures 17-21. In these graphs, the slope of a line connecting the data points increases with the amount of mercury adsorbed to the sediment. (The scales on the axes differ among the five figures.)

Adsorption on Cohansey Sand at pH 3.5 was similar for dissolved mercury and mercuric chloride, but much less for PMA (fig. 17a). At pH 5.5, adsorption was greatest for PMA, followed by mercuric chloride and dissolved mercury (fig. 17b). Adsorption on Bridgeton Formation sediments at pH 4.0 was greatest for dissolved mercury, followed by mercuric chloride and PMA (fig. 18a). At pH 6.0, however, adsorption was greatest for PMA (fig. 18b). Although PMA was the least adsorbed form of mercury on Cohansey Sand at low pH (3.5), it was the most adsorbed form on both Cohansey Sand and Bridgeton Formation sediments at pH 5.5-6.0.

Although there is considerable variation in the adsorption of dissolved mercury to the Cohansey Sand (fig. 19a), in general, adsorption at pH 3.5 was similar to that at pH 4.5, and was least at pH 5.5. This finding is consistent with results of other experimental work, in which adsorption of hydrated mercury ions decreased as pH increased above 4 (Barrow and Cox, 1992). Adsorption of dissolved mercury to Bridgeton Formation sediments (fig. 19b), in contrast, increased with increasing pH. Overall, a greater percentage of the dissolved mercury applied adsorbed to Bridgeton Formation sediments than to the Cohansey Sand at nearly equivalent pHs. (See appendix C for the calculated percent mass adsorbed.)

Adsorption of mercuric chloride as a function of pH followed a consistent pattern, with adsorption increasing with increasing pH for both sediment types (fig. 20a and b.). Overall, a slightly greater percentage of mercuric chloride applied adsorbed to Bridgeton Formation sediments than to Cohansey Sand. Results of the PMA batch experiments also followed a consistent pattern, in that more PMA adsorbed at high than at low pH (fig. 21a and b). The difference between adsorption characteristics at pH 3.5-4.0 and pH 4.5-5.0 was substantial for both sediment types. Overall, a greater percentage of the PMA applied adsorbed to Bridgeton Formation sediments than to Cohansey Sand.

Hogg and others (1978) found that, for loamy soils, adsorption of both mercuric chloride and PMA was greater to the soil with the greater organic-matter content. The sediments from the Cohansey Sand and the Bridgeton Formation have low organic-matter contents; thus, few sites on

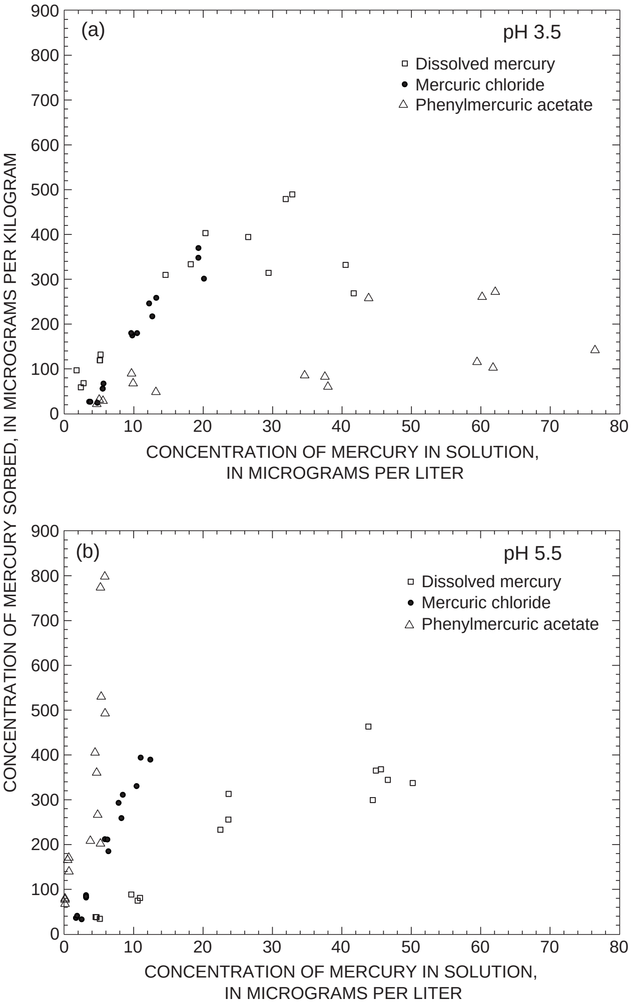

Figure 17. Adsorption of dissolved mercury, mercuric chloride, and phenylmercuric acetate on Cohansey Sand at (a) pH 3.5 and (b) pH 5.5.

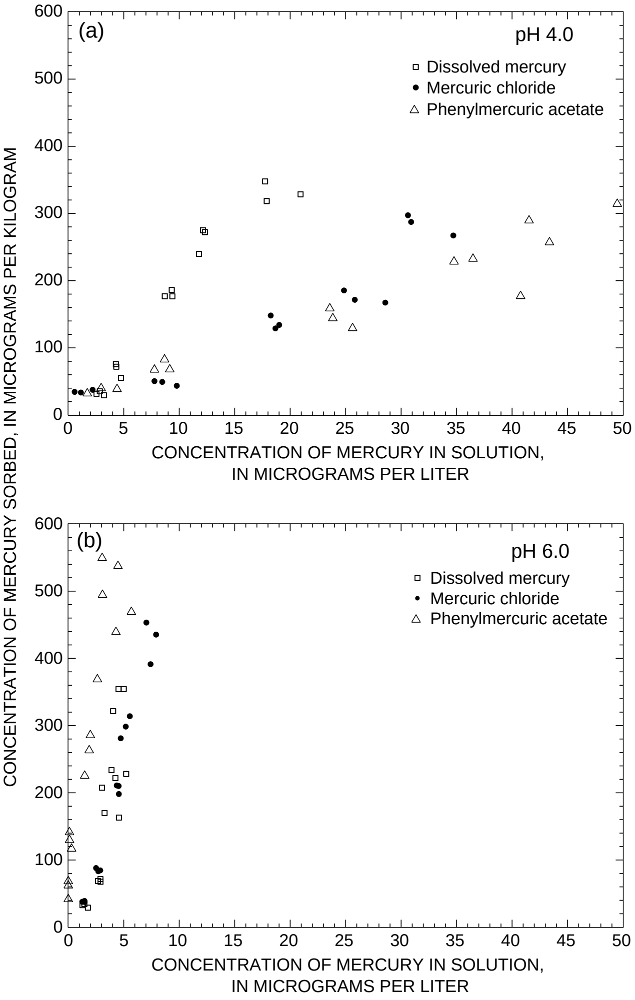

Figure 18. Adsorption of dissolved mercury, mercuric chloride, and phenylmercuric acetate on Bridgeton Formation sediment at (a) pH 4.0 and (b) pH 6.0.

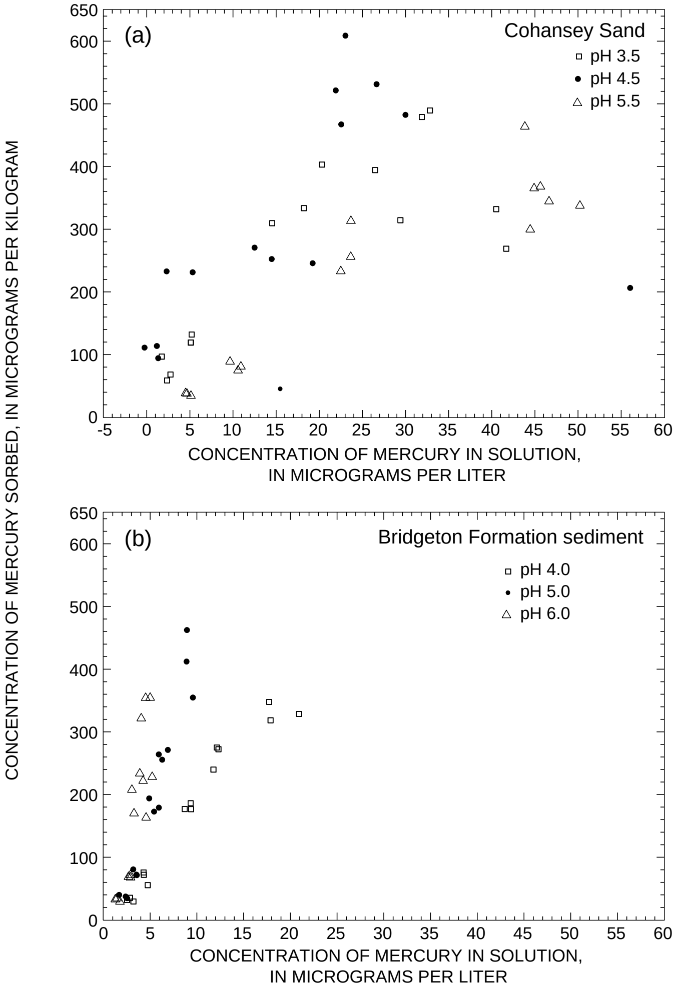

Figure 19. Adsorption of dissolved mercury on (a) Cohansey Sand at pH 3.5, 4.5, and 5.5, and (b) Bridgeton Formation sediment at pH 4.0, 5.0, and 6.0.

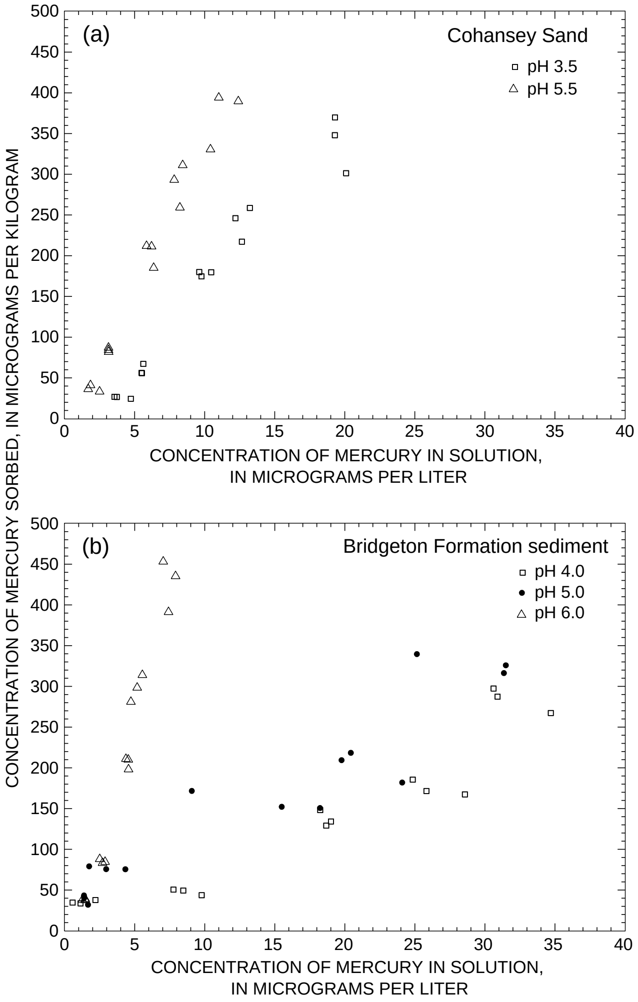

Figure 20. Adsorption of mercuric chloride on (a) Cohansey Sand at pH 3.5 and 5.5, and (b) Bridgeton Formation sediment at pH 4.0, 5.0, and 6.0.

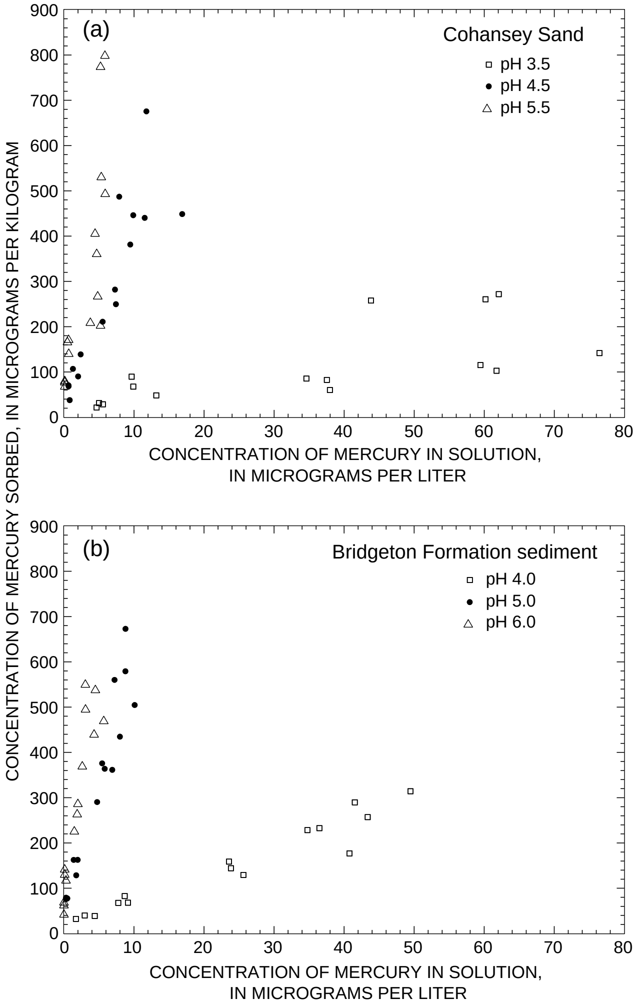

Figure 21. Adsorption of phenylmercuric acetate on (a) Cohansey Sand at pH 3.5, 4.5, and 5.5, and (b) Bridgeton Formation sediment at pH 4.0, 5.0, and 6.0.

organic matter are available for the adsorption of mercury. Organic matter was slightly more prevalent in the Bridgeton Formation sediments used in the experiments than in the Cohansey Sand and, therefore, the Bridgeton Formation sediment presumably had a slightly higher capacity for adsorbing mercury. Because the pHs in the experiments with Cohansey Sand were carried out at slightly lower pHs (3.5-5.5) than those in the experiments with Bridgeton Formation sediments (4.0-6.0), the results of the two sets of experiments cannot be compared quantitatively. Therefore, it is difficult to assess whether observed differences in the adsorption capacities of the two sediment types are a function of the sediment composition, an effect of differences in experimental conditions (pH), or both.

Geochemical results obtained by using the equilibrium model SOLMNEQ (Kharaka and others, 1988) indicate that dissolved mercury in water from sites 1, 4, and 10 is primarily in the forms $HgCl_2^{o}$, $HgCl_3^{-}$, and, in lesser concentrations, $HgCl^{+}$ and $Hg(OH)_2^{o}$ at the pHs measured in ground water. (Experimental pHs were similar.) Concentrations of other species probably are negligible. In the experiments, mercuric chloride was expected to be present predominantly as $HgCl_2^{o}$ and dissolved mercury was expected to be present predominantly as either $HgOH^{+}$ or $Hg(OH)_2^{o}$, depending on pH.

The iron hydroxide coatings on the quartz sands and silts of the Cohansey Sand and Bridgeton Formation could provide sites for adsorption of hydrated-mercury species; therefore, the amount of iron hydroxide present in the sediments used in the experiments and presence or absence of chloride also may explain observed differences in adsorption of mercury. Results of previous studies of mercury adsorption to soils and (or) sediments conflict with regard to the effect of iron hydroxide content. Semu and others (1986) found a negative correlation between mercury adsorbed and iron hydroxide (goethite) content; however, because these samples appear to have been ground and sieved, the coatings on soil grains that would provide adsorption sites could have been severely disturbed. Gunneriusson and Sjoberg (1993) found that mercury tended to be adsorbed to goethite in a freshwater environment with a high iron content, and that competing chloride ions inhibited adsorption. Lumsden and others (1995) found that slightly more mercury was adsorbed at lower pH to a soil with a greater organic-matter content than to a soil with greater extractable-iron content, and chloride ion reduced mercury adsorption to both soils. Nevertheless, Chandrajith and others (1995) found that mercury, in the absence of hydrogen sulfide, tended to be associated with hydrated iron oxides in sediments of the brackish Lake Jinzai. Schuster (1991), in an overview of various mercury studies, suggests that organomercurial compounds tend to be adsorbed to inorganic colloids whereas inorganic mercury compounds are more strongly bound to organic matter. This latter observation may explain some of the differences observed in the adsorption experiments reported here.

If mercury does adsorb to iron hydroxide coatings on the Cohansey Sand and the Bridgeton Formation sediments, the mechanism is likely to be specific adsorption rather than ion exchange. This chemisorption of metals, whereby the metal ions form hydroxide bridges with oxide or hydroxide surfaces, can involve either cationic or anionic species. The precipitation of iron hydroxides on the sediments of the Cohansey Sand is facilitated by iron-oxidizing bacteria (Means and others, 1981), and appears to be an ongoing process. Therefore, fresh hydroxide surfaces could become available over time--thereby increasing the capacity of the sediments to adsorb mercury unless some mobilizing agent is present in the aquifer to prevent adsorption. The hydroxides also could acquire coatings of organic materials to which the mercury could adsorb, possibly by forming complexes with various functional groups present in the organic matter.

Results of the adsorption experiments indicate that PMA is less strongly adsorbed than the inorganic species of mercury present in the solutions at pHs of 3.5 to 4.0. PMA is expected, however, to degrade relatively rapidly to produce mercuric ion and elemental mercury vapor in soils (Kimura and Miller, 1964); therefore, any inputs of mercury as PMA to the land surface where soil pHs are low might be expected initially to be more mobile than inorganic species, but ultimately, upon degradation to elemental mercury, to volatilize and (or) adsorb more strongly to sediments.

The distribution coefficients for mercury on the Cohansey Sand and Bridgeton Formation sediments were determined by using the Freundlich equation where n = 1. The distribution coefficients (K_d) determined from the results of the adsorption experiments are shown in table 11. The distribution coefficients were calculated by using mean values for each set of triplicate batch experiments. The values of K_d are relatively large, indi-

Table 11. Distribution coefficients (K_d)[1] for adsorption of mercury on sediments from the Cohansey Sand and the Bridgeton Formation

[Hg, dissolved mercury; $HgCl_2$, mercuric chloride; PMA, phenylmercuric acetate; --, no data]

Sediment	Hg species	pH3.5	pH4.5	pH5.5
Cohansey Sand	Hg	12.1	15.6	8.3
	$HgCl_2$	17.3	--	33.1
	PMA	3.06	42.0	93.0
		pH4	pH5	pH6
Bridgeton Formation	Hg	18.7	39.7	54.5
	$HgCl_2$	7.80	10.8	53.2
	PMA	6.14	63.5	120

[1] K_ds were calculated by averaging results for each set of three batch experiments at a given Hg concentration, and fitting a line to $K_d = \frac{Csc}{C}$, where Csc = concentration sorbed, corrected for pore fluid, and C = concentration in eluent. The line was constrained to pass through the origin. Units of K_d are liters per kilogram. Concentration data are presented in appendix C.

cating that mercury is expected to adsorb to aquifer sediments, or, by extrapolation, to the sandy soils developed on these sediments, as indicated by Yin and others (1996). The K_ds, however, are applicable only to sediments in the geochemical environments of the experiments; introduction of other constituents could change the adsorption capacity of the sediments. The presence of elevated mercury concentrations in ground water, accompanied by elevated concentrations of other constituents associated with human activities, indicates, however, that mercury has not remained adsorbed to in situ soils or sediments, but has been mobilized, as a solute, by adsorption to mobile colloids, or both.

Desorption from Aquifer Sediments

It was assumed during this study that mercury could desorb from soils, move downward, and adsorb to aquifer sediments. Therefore, preliminary column experiments were performed to test the effects of various infiltrating solutions on mercury adsorbed to aquifer sediments. The purpose of the experiments was to determine which solutions had the potential for removing substantial amounts of mercury from the sediments, and which did not. It also was assumed that the results would be grossly applicable to Coastal Plain soils. The compositions of the leaching solutions were designed to determine whether the nitrate and chloride present in elevated concentrations in ground water with elevated concentrations of mercury are involved in mobilizing mercury or merely are evidence of anthropogenic influences on the water chemistry. Several leaching solutions containing nitrate ion and a sodium chloride solution were used. The results of these preliminary leaching experiments are not rigorously comparable because the mass of mercury adsorbed to the sediments and the volume of leaching solution used differed among the experiments.

Results of previous analyses of mercury in New Jersey Coastal Plain soils and aquifer sediments in both undisturbed and disturbed settings indicated that the mercury content generally ranged from about 6.6 to 150 μg/kg (Barringer and others, 1997). The disturbed soils were sampled in residential areas at 6 of the 34 sites of elevated concentrations of mercury in ground water; these tended to contain less mercury than soils from two undisturbed forest areas, where the mercury concentrations were largest in organic and clay-rich horizons (Barringer and others, 1997). Aquifer sediments from a core at one site contained mercury in concentrations similar to those found in the soils. The largest mercury concentration found in soil or aquifer sediments was 258 μg/kg in a clayey silt from a soil- and aquifer-sediment core collected at the site of former thermometer factory (Barringer and others, 1997).

The first leaching experiments in which uninoculated sediment was used showed that the mercury concentrations were so small that any amounts removed by the leaching solutions in successive aliquots generally were not detectable. Because the mercury content of the sediments generally was small, it was decided that the sediments used in the column experiments needed to be inoculated in order to replicate a contaminated medium and to ensure that measurable amounts of mercury would be present in the effluent.

The inoculant solution was elemental mercury dissolved in 0.1-percent nitric acid that was diluted with distilled deionized water to give a concentration of 1,000 μg/L. As in the adsorption experiments, it was presumed that the mercury was ionized, and that some part of the mercury was present as hydrated mercury ionic species.

Each of the preliminary column experiments used for measuring desorption began with an adsorption phase, whereby the inoculant (1,000-μg/L mercury solution) was added slowly to the column, and the amount of mercury remaining in the column was determined as the difference between the mass added and the mass removed in the aliquots of solution that drained from the column. The amounts of mercury already adsorbed to the sediment when collected undoubtedly varied from column to column, as the sediment added to the columns could not be expected to be completely homogeneous. The concentrations of mercury adsorbed to sediments in the columns as a result of inoculation typically were more than 1,000 μg/kg (ppb); therefore, the effects of variations in the amounts of mercury in the sediments before inoculation on the results of the experiments were considered to be negligible. In the adsorption experiments performed previously, only about 1 percent of the mercury mass was lost to container walls or to volatilization when the most concentrated mercury solution (100 µg/L) was used (Peterson, 1993). Therefore, it was assumed that mercury loss from a 1,000-µg/L solution would be minimal.

The molar concentration of the inoculant was about 50 times that used by Yin and others (1996) in adsorption experiments, and 10 times that of the most concentrated solution used in the adsorption experiments performed during this study. Inoculants used by Yin and others (1997) in their desorption studies were four and eight times as concentrated as the inoculant used in this study. Because data on the mercury content of aquifer sediments are limited, a concentration of inoculant that might represent some of the more elevated values found in soils was used for this study. For example, Frank and others (1976) report up to 390 µg/kg of mercury in sandy soils from apple orchards in Ontario where PMA had been used as a foliar spray. Much larger concentrations of mercury (2,500-455,000 µg/kg) have been reported for a golf green in New Hampshire where mercurial pesticides were used (Estes and others, 1973). These golf-green soils contained about five times the amount of organic matter found in the Bridgeton Formation sediments, but their organic-matter content was comparable to that of some study-area soils (Barringer and others, 1997).

The capacity of the sediments in the columns to adsorb mercury during inoculation varied; some of the variation probably was related to differences in composition, and some to the kinetics of adsorption associated with small variations in the rate at which the inoculant was applied. In media with large organic-matter contents, such as sludges and peats, adsorption of metals typically is rapid (30 minutes or less) (Sparks, 1989). In adsorption experiments, mercury adsorbed quickly to clays (10 minutes to 1 hour to reach equilibrium), but more slowly to sands, reaching maximum adsorption in 3 to 4 hours (Reimers and Krenkel, 1974). Results of adsorption experiments with Bridgeton Formation sediments from New Jersey indicate that most of the dissolved mercury (the same solution used in the column experiments) adsorbed in the first 30 minutes, but that equilibrium of the sediment with this solution was not reached until after about 14 hours (Peterson, 1992). Therefore, variations in (1) sediment composition, (2) the flow rate of inoculant, and (3) the length of time the columns were equilibrated after the inoculant had drained from the column could result in differing degrees of adsorption. Additionally, some mercury may have volatilized. Moreover, the small size of the columns used in the experiments (10 cm in length) would tend to magnify any differences in adsorption rate. Use of larger columns (which was not practical in the timeframe of these preliminary experiments) probably would result in greater adsorption of the inoculant mercury.

Mercury adsorption on sediments in many of the experiments was about 50 to 90 percent of the mass of mercury applied, but, overall, ranged from about 24 to about 100 percent of the mass of mercury applied (table 12; app. D). The mass applied ranged from 500 to 580 µg for most of the experiments; smaller masses (230-400 µg) were used in two experiments.

Although the columns were drained after inoculation, some inoculant probably remained in the sediment pores. On the basis of results of the adsorption experiments, the amount of liquid remaining was likely to be small, but not negligible, as the pore volume of the sediments in the columns was about 40 mL. Insofar as the percentage of pore space retaining liquid was not known, and mass data (column, column and sediment, column and sediment and liquid, and column and sediment after the liquid was decanted) were not collected for these experiments, any correction for pore liquid would have been crude, at best. Therefore, in all cases, the amount calculated to be adsorbed is probably high by about 3 to 5 percent.

Typically, larger masses of mercury were removed from the columns by the first two volumes (20 mL, each) of leaching solution than by subsequent volumes. Some of the mass in the first two volumes may have been inoculant left in the columns after draining and aging, and some may have been weakly adsorbed mercury that would have adsorbed more strongly over time. This effect

Table 12. Masses and percents of applied mercury adsorbed to and leached from aquifer sediments in columns A, B, and C by six leaching solutions

[Hg, dissolved mercury; µg, micrograms; mL, milliliters; M, molar; values have been rounded]

Experimental conditions and results	Leaching solution and column								
	1 x 10^{-4} M Nitric acid			1.4 x 10^{-4} M Nitric acid			1 x 10^{-2} M Sodium chloride/water		
	A	B	C	A	B	C	A	B	C
Hg applied (µg)	520	500	500	360	400	400	240	230	230
Hg adsorbed (µg)	171	143	120	322	354	337	240	230	230
Hg adsorbed (percent)	32.9	28.6	24.1	89.4	88.4	84.1	99.9	99.9	99.9
Solution applied (mL)	600	600	600	640	600	600	100/390	100/310	100/390
Hg leached (µg)	50.0	44.0	47.1	240	173	297	132	159	187
Hg leached (percent)	29.2	30.7	39.1	74.5	49.1	88.4	55.2	69.3	81.5

Experimental conditions and results	Leaching solution and column								
	20-20-20 Fertilizer			1.4 x 10^{-2} M Sodium nitrate			1.4 x 10^{-1} M Sodium nitrate		
	A	B	C	A	B	C	A	B	C
Hg applied (µg)	520	520	520	560	580	560	360	520	520
Hg adsorbed (µg)	259	260	325	363	312	341	322	352	365
Hg adsorbed (percent)	49.8	50.0	62.5	64.8	53.7	60.9	89.3	67.8	70.2
Solution applied (mL)	580	580	580	1,000	1,000	1,000	350	350	350
Hg leached (µg)	413	511	517	54.6	39.8	16.9	57.4	67.0	71.7
Hg leached (percent)	160	197	159	15.1	12.8	4.97	16.4	19.0	19.7

was observed in all columns; the cause is not known. Because some of the mass of mercury removed may be from residual inoculant, the amount calculated to be leached from each column probably tends to slightly overestimate the amount of adsorbed mercury actually removed.

Total mercury was measured in the eluent from the desorption experiments. Because the experiments were not designed to determine the form of the mercury that was mobilized, it is not known conclusively whether the mercury measured in the eluent was desorbed and completely in dissolved form, or whether mercury-bearing colloids were released as well. Insofar as the eluent tended to be clear in the experiments reported, it is likely that colloids generally were not present in large amounts. Nonetheless, mercury that is removed from soils and aquifer sediments may, in some cases, be mercury adsorbed to fine clay, organic colloids, or iron and aluminum hydroxide colloids. Changes in ionic strength induced by infiltrating solutions could determine whether a colloidal suspension or a floc forms or persists. It is possible that the mechanism by which mercury is mobilized differs among experiments and, depending on the geochemical environment, among actual situations in the field.

Leaching Experiments Using Dilute Nitric Acid.--Nitrate concentrations typically were higher than expected background values in groundwater samples from sites of elevated concentrations of mercury in ground water. Consequently, solutions containing nitrate ion were used in some of the leaching experiments to determine whether the presence of nitrate could account for the mercury mobility, or whether it was an indication of anthropogenic effects rather than a causal agent. Several of the experiments were designed to examine the effects of added acidity, such as might be induced by acid rain, on mercury mobility. The role of acidic precipitation in mobilizing metals has been a topic of research during the past 2 decades (for example, Driscoll, 1980), and relations between mercury mobilization and acidic precipitation have been suggested (Brosset, 1977; Wood, 1980).

Acid rain contains both nitric and sulfuric acids. Because only nitric acid was used in the preliminary column experiments, the effect of the sulfuric acid component (which has decreased recently in some developed countries (Smith, 1988; Davies, 1988; McColl, 1988)) was not examined. Use of a nitric acid leaching solution also addresses effects of oxidation of ammonia (nitrification) from fertilizer or septic-system effluent because the oxidation of ammonia to nitrate produces hydrogen ion. Thus, acidic water with elevated nitrate concentrations can be produced by nitrification.

The leaching solution used in three columns (A, B, and C) packed with Cohansey Sand consisted of 1.0 x 10^{-4} M HNO_3 at a pH of 4.0. This pH is consistent with the low pHs measured in precipitation in southern New Jersey during the mid-1980's (Lord and others, 1990). The adsorption curves for this first set of columns (app. D) indicate that adsorption generally was no longer occurring when inoculation ceased--that is, the sediment in the columns had approached equilibrium with the inoculant. The amounts of mercury adsorbed from the inoculant were similar in all three columns, ranging from about 24 to about 33 percent of the mercury input (table 12; app. D). Because some of the inoculant was retained in the sediment pores, the actual amount adsorbed is likely to be slightly less.

A second leaching experiment using Cohansey Sand overlain by Bridgeton Formation sediments was carried out at a pH of 3.85 with a solution of 1.4 x 10^{-4} M nitric acid. Mercury input was smaller than in the previous acid experiment (table 12). Columns A, B, and C adsorbed from about 84 to 89 percent of the mercury input. The adsorption curves (app. D) for these columns indicate that adsorption was still occurring when inoculation ceased.

The results of the 1 x 10^{-4} M nitric acid leach of mercury adsorbed to Cohansey Sand indicated that from about 29 to 39 percent of the mercury applied to the sediments was removed (fig. 22a; table 12). A larger percentage of the smaller amount of applied mercury was leached by the 1.4 x 10^{-4} M nitric acid solution (fig. 22b; table 12).

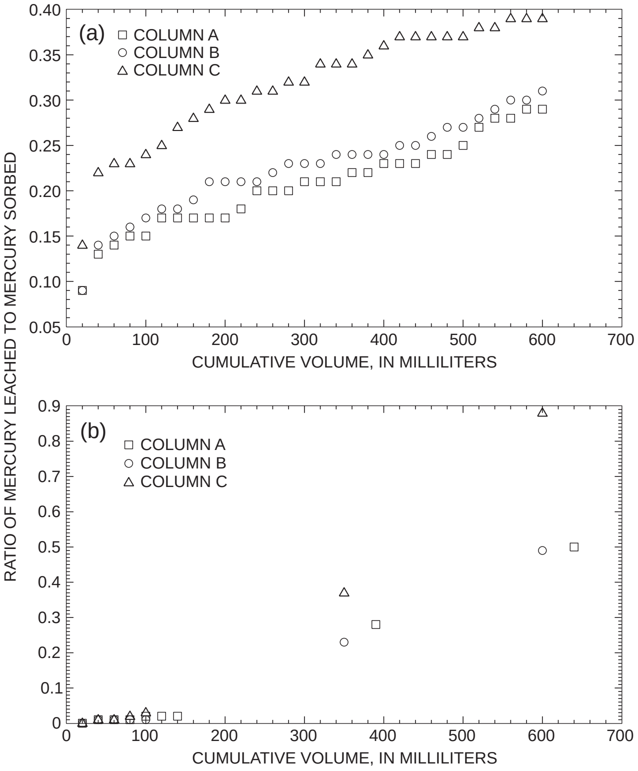

Figure 22. Elution of mercury (a) from three columns containing Cohansey Sand using 1 x 10^{-4} M nitric acid solution, and (b) from three columns containing Cohansey Sand overlain by Bridgeton Formation sediment using 1.4 x 10^{-4} M nitric acid solution.

The rates at which mercury was removed differed from one experiment to the other. The mass of mercury removed by the 1.4 x 10^{-4} M nitric acid was substantially greater than that removed by the 1 x 10^{-4} M nitric acid. Although the more concentrated acid solution may have been responsible in part for the increased mass of mercury removed, other factors, such as a difference in the length of time the columns were aged after inoculation, may account for some of the difference in results.

Because the concentration of nitrate in the leaching solutions used in these preliminary experiments was small, it is unlikely that nitrate ion formed mobile complexes with the mercury. Previous research to determine species of mercury in chloride and nitrate solutions (Gilmour, 1971) indicates that the presence of mercury-nitrate complexes generally is negligible; therefore, the principal species in the experimental nitrate solutions most likely were mercury cations and hydroxide species.

Leaching Experiments Using a Sodium Chloride Solution.--A preliminary column experiment was carried out to determine the effect of infiltrating chloride solution on adsorbed mercury in aquifer sediments. Runoff containing road salt is capable of releasing metals from soils (Bauske and Goetz, 1993; Amrhein and others, 1992; Feick and others, 1972). Additionally, sodium and chloride are constituents of domestic sewage, in which chloride concentrations typically range from 30 to 100 mg/L (Miller, 1980, p. 218). Furthermore, as discussed earlier, the potential for formation of mercury chloride complexes is well-documented.

Three columns (A, B, and C) packed with Cohansey Sand overlain by Bridgeton Formation sediments were inoculated with 230 or 240 µg of mercury; the adsorption curves (app. D), which are virtually identical for each of the three columns, indicate that adsorption was continuing when inoculation ceased. Thus, more mercury could have been adsorbed. A 1 x 10^{-2} M NaCl (100 mL) solution, followed by distilled water, was used to leach the columns; these solutions removed from 55 to 82 percent of the mercury adsorbed to the sediments (fig. 23; table 12). Nearly all of the mercury removed was leached by the sodium chloride solution; the distilled water removed only another 2 to 6 percent of the applied mercury. Formation of mercury chloride complexes is the most likely mechanism for removing mercury from these columns.

Leaching Experiments Using Fertilizer Solutions.--A preliminary column experiment was designed to illustrate the effect of a solution containing commercial chemical fertilizer (20 percent N, 20 percent P, 20 percent K) on mercury mobilization. Of the 520 mL of inoculant applied to three columns, with a mass of 520 µg, about 50 to 62 percent of the applied mercury was adsorbed. Measurements of mercury in the eluent during the leach with the 20-20-20 fertilizer solution indicate that not only was 100 percent of the applied mercury removed, but additional mercury was eluted from the columns. The total amount of mercury that was eluted ranged from 159 to 197 percent of the amounts that had adsorbed to the sediment from the inoculant (fig. 24; table 12).

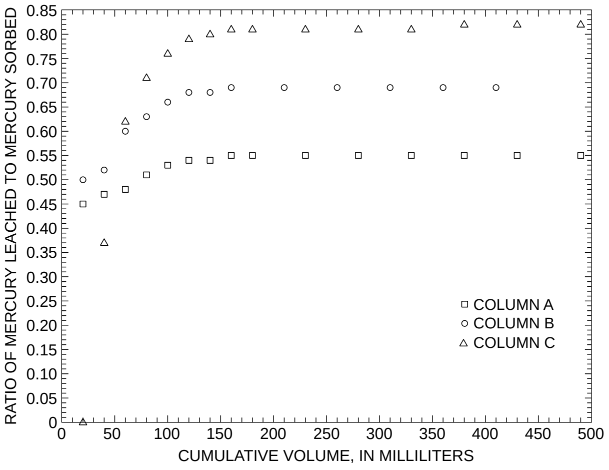

Figure 23. Elution of mercury from columns A, B, and C containing Cohansey Sand overlain by Bridgeton Formation sediment using 1 x 10^{-2} M sodium chloride solution and distilled water.

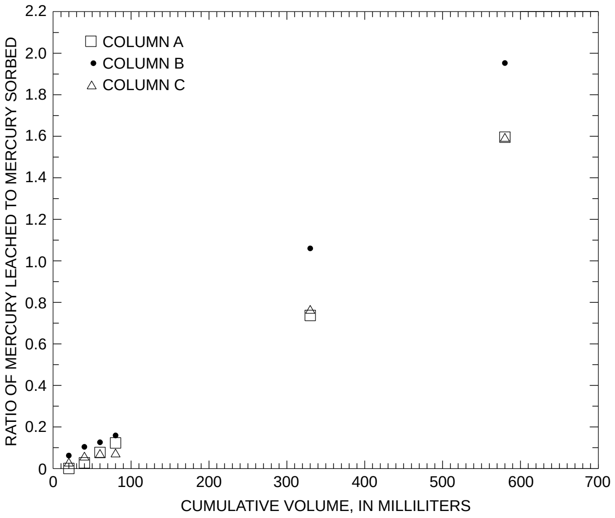

Figure 24. Elution of mercury from columns A, B, and C containing Cohansey Sand overlain by Bridgeton Formation sediment using solution containing 20-20-20 fertilizer.

The 20-20-20 fertilizer solution was found to contain about 280 µg/L of mercury. Mercury in commercial fertilizers has been reported at various concentrations: 4 to 47 ng/g (or µg/kg) were measured by Frank and others (1976); Kabata-Pendias and Pendias (1992) report 10 to 120 µg/kg. Superphosphate fertilizers in particular tend to contain high concentrations of mercury (up to 3,100 ng/g (Andersson, 1979)). The mercury can be contributed by the phosphate rock used, wherein the mercury concentrations can vary widely. Jones and Hinesly determined a mean concentration of 177 µg/kg in five samples; Kabata-Pendias and Pendias (1992) report up to 1,000 µg/kg in phosphorite. Another reported source of mercury and other metals in fertilizers is unregulated wastes recycled into fertilizers (Savitz, J.D., Hettenbach, Todd, and Wiles, Richard, 1998, Factory farming--Toxic waste and fertilizer in New Jersey, 1990-1995: Factory farming, March 26, 1998, Environmental Working Group, Washington, D.C., 11 p., accessed May 26, 1998, on the World Wide Web at URL http://www.ewg.org.).

Because similar results were obtained in later experiments in which analyses were done by CVAAS, no interference from other elements is suspected in the ICP analytical results for the preliminary 20-20-20 column experiment. Of the 413 to 517 µg of mercury removed from the columns (table 12), between 259 and 325 µg would have been mercury adsorbed from the inoculant, and the rest (154-251 µg) was either already adsorbed to the sediment when it was put into the column, or was in the leaching solution. Compared with mercury concentrations measured in soils and aquifer sediments during the study, 154 µg on about 100 g of soil (about 1,540 µg/kg) is a large concentration. Therefore, it is unlikely that the extra mercury in the column effluent was derived solely from mercury already present in the sediment. The standards for the experiment were mixed in 20-20-20 solution and then zeroed to correct for the mercury present in the leaching solution. If this did not correct for the mercury in the leaching solution, then the 580 mL of 20-20-20 leaching solution, which contained an average of 162 µg of mercury, may have been the source of the additional mercury in the system.

Whatever the reasons for the anomalous results obtained during the 20-20-20 fertilizer experiment, it is clear that this solution can remove adsorbed mercury. Concentrations of ammonium, nitrate, phosphate, and potassium (and other constituents not reported on the label) were not measured in the effluent because the analytical instruments for these analyses were not available at the time. If mercury was removed by ion exchange, then concentrations of the ion replacing it would be smaller in the effluent than in the influent. Additional column experiments would be needed to determine the mechanism involved.

Sodium nitrate, which promotes a decrease in soil acidity, is sometimes used as a fertilizer (Foth, 1951, p. 347). A solution of 1.4 x 10^{-2} M sodium nitrate was used in a set of leaching experiments, which were carried out at an initial pH of 5.16. The mass of mercury adsorbed ranged from about 54 to 65 percent of the amount applied. The sodium nitrate solution removed only about 5 to 15 percent of the mercury calculated to be adsorbed to the sediment (fig. 25a; table 12). The 20-20-20 fertilizer solution was calculated to be about 3 x 10^{-3} M in nitrate, and it was by far the more effective leaching solution. In a second set of leaching experiments with sodium nitrate, a 1.4 x 10^{-1} M solution was used. About 68 to 89 percent of the mercury input to the columns adsorbed, and about 16 to 20 percent of the amount adsorbed was removed (fig. 25b; table 12).

Similar masses of mercury were adsorbed in the two sets of sodium nitrate experiments, but a smaller volume of the concentrated solution removed more mercury than a greater volume of the dilute solution. Therefore, the ionic strength of a solution may be a factor in leaching mercury. The dramatic difference in the degree of mercury mobilization by the 20-20-20 fertilizer solution compared with the two sodium nitrate solutions could, therefore, be attributable to differences in ionic strength; alternatively, the difference could result from the presence of species or compounds other than nitrate. If the 20-20-20 fertilizer solution contained ethylenediaminetetraacetic acid (EDTA), as do some commercial fertilizers, mercury may have been chelated by the EDTA and become mobile.

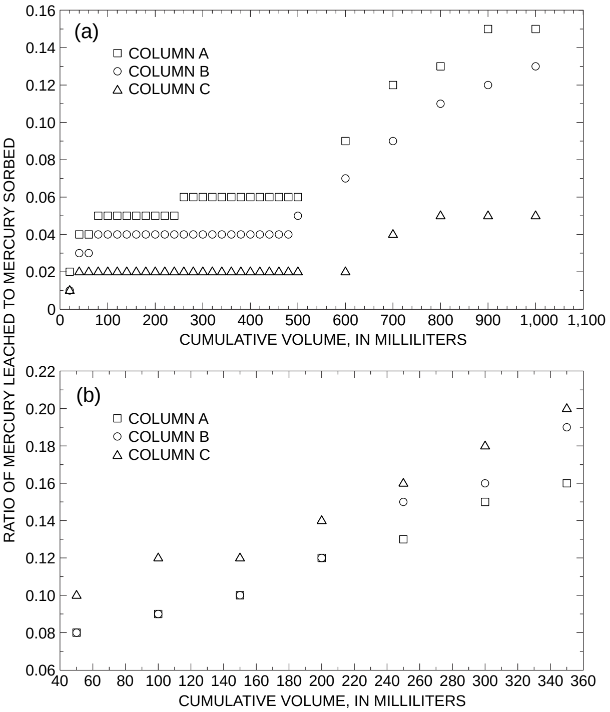

Figure 25. Elution of mercury (a) from three columns containing Cohansey Sand using 1.4 x 10^{-2} M sodium nitrate solution, and (b) from three columns containing Cohansey Sand using 1.4 x 10^{-1} M sodium nitrate solution.

Given the relative ineffectiveness of the sodium nitrate solution at removing mercury, it appears that sodium ion participates only minimally in the desorption process. These results are consistent with those reported by Behra and Bourg (1987), whereby sodium ion had no significant effect on mercury retention by sediments. Overall, results of the preliminary column experiments appear to indicate that nitrate ion is not very effective at removing mercury, and that its presence in ground water in elevated concentrations probably is only an indication of anthropogenic effects on the water chemistry. Of the constituents in the 20-20-20 fertilizer, the influence of phosphate cannot be discounted, but this anion typically is adsorbed to soils and is, therefore, unlikely to reach the water table if it is input at the land surface. Phosphate is introduced in the subsurface, however, in septic-system effluent. Whether phosphate, potassium, or ammonium (or EDTA and (or) chloride, if present) is the effective mobilizing agent in the 20-20-20 fertilizer has yet to be determined.

Although the results of the adsorption experiments indicate that much of the mercury applied to aquifer sediments from the Cohansey Sand and the Bridgeton Formation is adsorbed, the results of the column leaching experiments show that several different solutions that are related to human activities can release some mercury from the aquifer sediments. On the basis of previous investigations and current knowledge of mercury chemistry, a sodium chloride solution could be predicted to promote mercury mobility. The preliminary results achieved by using the 20-20-20 fertilizer, which was effective at moving mercury through the sediments in the column, are less predictable. Furthermore, the fertilizer also contains mercury, and could constitute yet another source to study-area soils and, perhaps, to ground water in agricultural or residential settings.

Conditions and Possible Mechanisms for Mobilization of Mercury

Persistence of Mercury in Soils

In order for mercury applied to soils to leach, it must persist in the soils after application. Because elemental mercury is highly volatile, and some mercury compounds are moderately volatile, large amounts of mercury potentially could vaporize from soils to which it is applied. Such vaporization evidently is not significant in southern New Jersey, if the hypothesis that land-applied substances are a source of elevated mercury concentrations in ground water is correct.

Estimates and measurements of the amount of volatilization of mercury compounds, such as pesticides, vary, and variability in volatilization should result in variability in the amount of mercury retained by soils. Experiments conducted by Poelstra and others (1974) indicate that evaporation of mercuric chloride from sandy soils was slow (less than 1 percent of the mercuric chloride added over a period of about 300 days). Hogg and others (1978) ascribe an 80.6-percent recovery of mercuric chloride applied to a sandy soil (86 percent sand) to volatilization. Rogers (1979) applied mercuric chloride, mercuric nitrate, mercuric acetate, and mercuric oxide solutions to a sandy loam at a concentration of one part per million (1,000 µg/kg), and measured 38-, 36-, 26-, and 20-percent losses of mercury, respectively, from these compounds. Rogers and McFarlane (1979) found that 43 percent of mercury applied as mercuric nitrate was lost from a sandy loam soil within the first 6 days of the experiments, which they ascribe to mediation “by a biological system.”

Many of the soils used in the experiments cited appear to be similar to New Jersey Coastal Plain soils. Given the variable nature of the experimental results, however, it is difficult to determine how much applied mercury could be expected to evaporate from study-area soils, and under what conditions. Although considerable data are available for mercury evaporation in experimental systems, data regarding evaporation of mercury from farm fields are limited. Hence, any extrapolation from the laboratory results to the field situation needs to be done with caution, because the systems may not be comparable.

Little volatilization would be expected if a mercury source were buried; such a source could include discarded pesticide containers, refuse from thermometer manufacture (a home industry scattered throughout much of southern New Jersey in the early to mid-20th century), and discarded paints. The existence of such dumps is alluded to by some local residents, but locations typically are not known. Buried mercurial debris could represent a small but concentrated source of mercury that is highly likely to persist.

Some measurements of mercury in soils, however, indicate accumulation and persistence of anthropogenic mercury. Measurements of mercury in undisturbed forest soils during the initial phase of this study (Barringer and others, 1997) indicate that atmospherically deposited mercury does accumulate, at least to concentrations of 150 µg/kg. Frank and others (1976) report that mercury contents in apple orchards in Ontario where PMA was used were “slightly elevated.” Because the soil samples analyzed for mercury were oven-dried before digestion (Frank and others, 1976, p. 184), the measured mercury concentrations, which were as high as 1,140 µg/kg, could be less than the actual mercury content of the soils. Results of other studies summarized by Andersson (1979) indicate that cultivated soils (particularly those where rice is grown) and deltaic soils can contain from several hundred to more than 10,000 µg/kg of mercury. Estes and others (1973) measured mercury that had accumulated in a golf-course soil where organomercurial pesticides had been applied over a period of 15 years at a rate of 2.1 kg/ha (1.8 lb/acre). The mean concentration of 12 measurements made from the surface to a 1-ft depth was 220 ppm (2.2×10^5 μg/kg), and the median was 270 ppm; measurements ranged from 2.25 to 455 ppm. These mercury concentrations indicate that mercury from land-applied substances can accumulate and persist substantially in some soils, despite any loss through volatilization.

The mercury concentrations measured by Estes and others (1973) are more than three orders of magnitude greater than those measured in the samples of New Jersey soils collected during the present study. The samples collected during this study, however, were mostly from disturbed soils in the residential areas where ground water contains elevated concentrations of mercury. Thus, if the residential soils originally contained mercury in large concentrations, it may have already been leached from the soils, insofar as the elevated concentrations in the ground water are much greater than those that would be expected to occur naturally. Overland runoff probably removes little mercury from these soils because runoff is negligible under most conditions in the highly permeable sandy soils of the New Jersey Coastal Plain.

Geochemical Mechanisms of Mobilization

Results of many previous studies of mercury mobility in soils (summarized by Schuster, 1991) indicate that most applied mercury remains in surface soil horizons, typically is associated with organic matter, and seldom is leached. In order for anthropogenic mercury to reach depths within the aquifer where it is tapped by domestic wells, physical and (or) chemical processes that promote desorption or otherwise remove mercury from soils and aquifer sediments and transport it to depth within the aquifer must occur. Results of two recent studies (Romken and others, 1996; Temminghoff and others, 1997) of interactions among organic carbon, copper, and calcium, however, point to another mobilizing possibility. Copper, like mercury, binds to organic matter, which can complex with calcium and coagulate as a semisolid. In the absence of calcium, organic matter is solubilized and copper is mobilized. Liming of agricultural soils would tend to keep organic matter as a solid; cessation of liming would promote the presence of dissolved organic matter, which is mobile and, therefore, a vehicle for moving metals such as copper and mercury from soils to ground water. Whether soluble organic matter bearing complexed mercury redeposits on iron hydroxide coatings on sand grains is not known. The concentrations of mercury and iron in water samples from borehole 1 (discussed earlier) indicate a possible association of mercury with iron hydroxide in the aquifer.

The mineral phases in study-area soils and sediments to which the mercury is adsorbed have not been conclusively determined. Mercury in the aquifer sediments may be adsorbed to hydrated iron oxide coatings on sand grains (Peterson, 1992); evidence of this finding is consistent with results of a study of mercury in lacustrine sediments (Chandrajith and others, 1995). If iron hydroxide coatings are the principal adsorbers of mercury in the aquifer, mercury may become mobile by more than one mechanism. These can include complexing of adsorbed mercury, ion exchange on the hydroxide surface, reduction and solubilization of the iron through changes in pH and redox conditions resulting in release of mercury, and creation of mobile colloids that carry adsorbed mercury.

Colloids can be created and mobilized by chemical and physical means. In addition to the stirring of fine particles by well pumping, the introduction of chemicals into soils and the aquifer can mobilize colloids. Ryan and Gschwend (1994), in experiments with sediment from the New Jersey Coastal Plain, found that surfactants are effective in mobilizing colloids. The colloids are mobilized when electrostatic repulsion between surfaces of clays and iron oxides increases; the surfactants promote the repulsion, presumably as phosphate and sulfonate functional groups form complexes with oxides and reverse the surface charge.

Mechanisms Associated with Land-Use Change

Activities associated with residential development can have a profound effect on physical and geochemical processes in soils and sediments of both the unsaturated and saturated zones. In a change from agricultural to residential land use, soils may no longer be limed, and the mobility of mercury may be enhanced by the solubilization of organic matter. With any change from agricultural (or undeveloped) to residential land use, soil is disrupted more substantially by development than by agricultural activities; roads are built, and use of road salt is likely to increase. Pumping from newly installed wells in previously undisturbed areas of the aquifer and surfactants in wastewater from new septic systems may mobilize colloids.

Available data are insufficient to determine the importance of the physical disruption of the soils in the processes that move mercury from the land surface to the ground water. Plowing might be expected to move mercury from the surface deeper into the soil column; simulation of the migration of solutes as a result of plowing indicates that this could occur (Schimmack and Bunzl, 1986). In some studies of cultivated soils mercury concentrations were largest in surface horizons, whereas in others mercury was evenly distributed or increased with depth in the soil profile (Andersson, 1979). If soils at a site undergoing residential development contain elevated concentrations of mercury, the mechanical stirring produced by earth-moving activities could move mercury adsorbed to surficial soils deeper within the soil column, and, therefore, closer to the water table. Most of the sites where elevated concentrations of mercury in ground water have been identified are housing developments. Only 2 of the 34 sites include domestic wells located in agricultural settings for which no detailed history of activities is available.

Although none of the preliminary experiments specifically addressed the effects of septic-system effluent, many of the constituents of fertilizers and road salt that potentially could affect mercury mobility, such as urea, ammonia, potassium, and chloride, also are present in septic-system effluent. The nearly ubiquitous occurrence of chloroform in about 1,500 ground-water samples from sites in Atlantic County indicate that chlorine from septic-system effluent may have affected the water tapped by domestic wells in the residential neighborhoods. In addition, use of chlorinated solvents as septic-system cleaners may be the source of some of the VOCs measured in water from many wells at the 34 sites. The effect of these organic compounds on mercury mobility is not known with certainty, although the mobility of mercury can be enhanced in the presence of anthropogenic organic compounds (Niebla and others, 1976). Furthermore, the presence of anthropogenic organic compounds and (or) septic-system effluent can promote a reducing geochemical environment in which mercury mobility may be enhanced. The effects of a reducing environment on mercury mobility in flooded soils and lake sediments have been explored; mercury that is adsorbed to iron oxyhydroxide is released when the iron is reduced (Dmytriw and others, 1995). Gambrell and others (1980) found that moderately acid, reduced conditions increased the concentrations of soluble mercury in bay-sediment samples. Additional work is needed to assess definitively the effect of organic solvents and (or) changes in redox conditions on mercury mobility.

Because the total-mercury-concentration measurements were skewed toward residential areas where mercury contamination was found and, typically, many wells were sampled in a relatively small area, it is difficult to test conclusively the hypothesis that change in land use from agricultural or undeveloped to residential is a key factor in mobilizing mercury. The density of wells in areas of current agricultural or undeveloped land is substantially less than that in areas of residential land use; therefore, it is currently impossible to test whether the areal extent of mercury contamination of ground water in the residential areas is matched by that in agricultural or undeveloped areas. Total-mercury-concentration measurements in water from wells from areas of current agricultural and undeveloped land in New Jersey are few compared with those from wells in the residential areas. As an additional complication, many of the water samples from the wells in agricultural or undeveloped areas were filtered before analysis, so mercury concentrations in these samples may be smaller than would have been measured had the samples not been filtered. Therefore, the available data do not permit a thorough assessment of differences in mercury concentrations with land use because total- (unfiltered) mercury concentrations in ground water underlying agricultural or undeveloped areas may or may not be smaller than those measured in the unfiltered samples from domestic wells at the 34 sites.

The depths at which the wells containing elevated concentrations of mercury are completed indicate that mercury-bearing recharge probably moved into the aquifer several decades ago. Mercury concentrations above the MCL were found at depths ranging from 50 to 200 ft below land surface. Well-construction data were available for only 456 of the 2,239 wells at the 34 sites, but the distribution of well depths for the 456 wells is assumed to be representative of the total population of wells. Mercury concentrations that exceed background levels thus far have been found only in water affected by human activities and not in chemically pristine water. The occurrence of elevated mercury concentrations with elevated concentrations of generally conservative constituents like chloride and nitrate may indicate that mobilized mercury experiences little or no retardation. Consequently, if a mean depth to water of 19 ft (from data for the 167 wells for which water-level data were available) and vertical ground-water velocities of 2.3 to 3.3 ft/yr are assumed, then water containing mercury that is tapped by wells ranging from 50 to 200 ft in depth must have entered the saturated zone between about 9.4 and 79 years ago. The maximum estimated ages of the contaminated water in most of the wells, which are 50 to 150 ft deep, range from 13.5 to 57 years; these ages appear to coincide with the period in which land-use changes were occurring. The possible effects of pumping domestic wells on vertical flow velocities are not considered in these estimates because of a lack of data. Preliminary results of simulations with a finite-difference ground-water flow model indicate that pumping from small-volume domestic wells could affect the vertical distribution of water ages, particularly near ground-water divides (Shilpa Rajagopalan, University of Pennsylvania, oral commun., 1999).

Because about 20 percent of the 34 sites do not have a known history of agricultural land use as far back as the 1940’s to 1950’s, a scenario involving agricultural mercurial pesticide use accompanied by fertilizer and lime application may not be appropriate at these sites. Mercurial pesticides may have been used for lawn improvement and maintenance, as these pesticides were recommended at one time for use in controlling crab grass (Wolf and Engel, 1948), and have continued to be used on turf after their use on food crops was banned in 1972. Although one or more possible point sources of contamination, such as landfills, illegal dumps, military operations, and industrial and commercial enterprises, commonly are present within 3 mi of most of the 34 sites, an extensive survey of water-quality data from monitoring wells and assessment of local hydrology does not yet conclusively link any known point sources of mercury with the 34 sites. Several of these point sources, however, ultimately may be linked to a few of the sites as more data become available. Nevertheless, it appears that most of the mercury now found in ground water is contributed by non-point sources. Results of the desorption experiments indicate that any contribution of mercury by atmospheric deposition (which might include mercury outgassed from exterior house paint), as well as mercury contributed by other sources, can be mobilized by more than a single agent. Nevertheless, leaching of atmospherically deposited mercury by acid rain appears to be minimal, as mercury concentrations in undisturbed forest soils tend to be elevated relative to those in disturbed residential soils, and shallow ground water in forested areas does not appear to contain elevated concentrations of mercury (Barringer and others, 1997).

Results of the adsorption experiments performed during this study and the study of Yin and others (1996) indicate that mercury initially can be adsorbed to the soils and aquifer sediments of southern New Jersey. As indicated by the preliminary leaching experiments performed during this study and other experimental work (Behra and Bourg, 1987; Feick and others, 1972; Lumsdon and others, 1995; Romken and others, 1996), it also appears that mercury can be mobilized either by substances that are applied to the land surface and shallow subsurface, or by the cessation of application of some substances. In short, changes in the geochemical environment can cause mercury to become mobile.

Unanswered questions with regard to mercury mobility in the New Jersey Coastal Plain include (1) whether methylization of mercury, which could promote mercury mobility, occurs in the soils; (2) whether the VOCs measured in ground water from the same areas in which the elevated mercury concentrations are found have played a role in mobilizing mercury; (3) which compounds in fertilizer mobilize mercury, and which form or forms of mercury are mobile; and (4) whether occurrences of elevated mercury concentrations in ground water are related to land-use change, particularly change to residential land use and the attendant introduction and (or) increase of pumped wells and septic systems. Changes in the geochemical environment appear to be a key factor, but more than one geochemical process may be involved in mercury mobility.

Land use is the common thread that ties together the findings of the current study and the questions raised as a result of the study. It appears that substantial amounts of mercury could have been introduced to the environment by various activities associated with particular land uses. The data collected thus far indicate that activities associated with various land uses or changes in land use have provided mechanisms for mobilizing the mercury so that its ultimate destination is the drinking-water supply of southern New Jersey.

SUMMARY AND CONCLUSIONS

Water from 265 wells tapping the unconfined Kirkwood-Cohansey aquifer system in New Jersey contains concentrations of mercury that exceed the U.S. Environmental Protection Agency MCL of 2 µg/L. The wells are predominantly domestic wells in residential areas that were developed during the 1950's through the 1970's. In addition, of the 2,005 other wells sampled from 1982 through 1993, about 1,000 yielded water with concentrations of mercury greater than reported detection limits. The wells sampled generally are finished at depths from 50 to 200 ft from land surface.

Naturally occurring mercury concentrations in ground water from the Kirkwood-Cohansey aquifer system typically are less than 10 ng/L. On the basis of background concentrations, 34 sites of elevated mercury concentrations (1 µg/L or greater) in ground water were identified; mercury concentrations in water from one or more wells at each of 32 of the 34 sites exceeded the MCL. The residential areas generally were upgradient or far from possible point sources of contaminants, such as landfills, military bases, industrial operations, or commercial establishments. On the basis of estimates of the age of ground water as a function of depth from land surface and analyses of water samples for tritium and helium concentrations, ground water containing elevated concentrations of mercury appears to have been recharged between 9.4 and 79 years ago. Investigations of previous land use indicate that about 75 percent of the 34 sites of elevated mercury in ground water are on or adjacent to former agricultural land.

Although constituents other than mercury and VOCs were rarely measured in water samples from the 2,270 wells, examination of other water-quality characteristics of and constituents in water samples from wells and boreholes at 6 of the 34 sites reveals that nitrate concentrations commonly are elevated above background levels (which typically are undetectable at 0.01 mg/L), and, in some cases, exceed the MCL of 10 mg/L. Chloride concentrations also are elevated above expected background levels. Elevated mercury concentrations typically have not been detected in ground water in undeveloped areas, but are found where concentrations of nitrate and chloride are elevated.

The presence of elevated nitrate concentrations in ground water in conjunction with elevated magnesium, barium, and strontium concentrations may be the result of past inputs of agricultural chemicals at several of the sites of elevated mercury concentrations in ground water. Elevated nitrate concentrations also can result from oxidation of ammonia compounds in residential septic-system effluent. The presence of chloroform in water from many of the 34 sites may be attributable to septic-system effects on ground water, probably resulting from the long-term use of household bleach.

Batch equilibrium experiments were performed to determine the degree to which various mercury compounds adsorbed to the quartz-rich aquifer sediments. Preliminary column leaching experiments were initiated to illustrate the effectiveness of various compounds in removing adsorbed mercury from aquifer sediments. Batch equilibrium experiments at low to moderate pH (3.5-4.0, 4.5-5.0, and 5.5-6.0) were conducted to measure the sediment's ability to adsorb dissolved mercury (Hg), mercuric chloride ($HgCl_2$), and phenylmercuric acetate (PMA). The distribution coefficients determined for these sediments are moderately high, and mass balances calculated for the experimental systems indicate that, in nearly all cases, most of the mercury added to the sediments was adsorbed. The adsorption of the mercury compounds to the sediments at pH 5.5 and 6.0 decreased in the order $PMA>HgCl_2>Hg$ for the Cohansey Sand and $PMA>Hg>HgCl_2$ for the Bridgeton Formation sediments. At pH 3.5 and 4.0, PMA was the least adsorbed on both sediments, and the order of adsorption was $HgCl_2>Hg>PMA$ for the Cohansey Sand and $Hg>HgCl_2>PMA$ for Bridgeton Formation sediments.

Mercury was leached from inoculated aquifer sediments by using solutions of 1×10^{-2} M NaCl, 1×10^{-4} M HNO_3, 1.4×10^{-4} M HNO_3, a commercial fertilizer (20-20-20), and two different concentrations of sodium nitrate, also a fertilizer. The results of the 20-20-20 fertilizer experiment were anomalous in that more mercury was removed than had been applied to the sediments in the columns, apparently indicating that mercury adsorbed to the sediment in situ also was removed; the 20-20-20 fertilizer also contained mercury. The 1×10^{-2} M NaCl solution, simulating road salt runoff, was moderately effective in removing mercury. The more concentrated nitric acid solution also was moderately effective, but the more dilute nitric acid removed less mercury. The sodium nitrate solutions removed relatively little mercury from the sediments. The elevated concentrations of nitrate in ground water from some of the 34 sites most likely are the result of anthropogenic effects on ground-water quality and probably do not act as a mobilizing agent.

Land use at the 34 sites generally has changed from agricultural or undeveloped to residential. Ground-water-quality data for wells in agricultural areas indicate that agricultural chemicals have affected water quality, but the density of wells in these areas is too small to support or refute a hypothesis that agricultural land use results in elevated concentrations of mercury in ground water. Mercury-concentration data are skewed toward ground-water samples from residential areas where the number of wells in a relatively small area is great. Although use of agricultural chemicals is one of several possible sources of mercury to Coastal Plain soils, land-use change appears to be an important factor in the mobilization of mercury from soils or sediments to ground water. Additional work is needed to determine the roles of physical disturbance of soils, installation and pumping of domestic wells, and installation, use, and maintenance of septic systems in removing adsorbed mercury from soils and sediments and transporting it through the aquifer.

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