# Sources and Occurrence of Chloroform and Other Trihalomethanes in Drinking-Water Supply Wells in the United States, 1986–2001

By Tamara Ivahnenko and John S. Zogorski

Scientific Investigations Report 2006–5015

**U.S. Department of the Interior**
**U.S. Geological Survey**

**U.S. Department of the Interior**
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**U.S. Geological Survey**
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*Suggested citation:*
Ivahnenko, Tamara, and Zogorski, J.S., 2006, Sources and occurrence of chloroform and other trihalomethanes in drinking-water supply wells in the United States, 1986–2001: U.S. Geological Survey Scientific Investigations Report 2006–5015, 13 p.

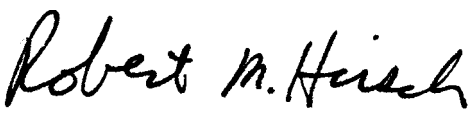
# FOREWORD

The U.S. Geological Survey (USGS) is committed to serve the Nation with accurate and timely scientific information that helps enhance and protect the overall quality of life, and facilitates effective management of water, biological, energy, and mineral resources. (http://www.usgs.gov/). Information on the quality of the Nation's water resources is of critical interest to the USGS because it is so integrally linked to the long-term availability of water that is clean and safe for drinking and recreation and that is suitable for industry, irrigation, and habitat for fish and wildlife. Escalating population growth and increasing demands for the multiple water uses make water availability, now measured in terms of quantity and quality, even more critical to the long-term sustainability of our communities and ecosystems.

The USGS implemented the National Water-Quality Assessment (NAWQA) Program to support national, regional, and local information needs and decisions related to water-quality management and policy. (http://water.usgs.gov/nawqa/). Shaped by and coordinated with ongoing efforts of other Federal, State, and local agencies, the NAWQA Program is designed to answer: What is the condition of our Nation's streams and ground water? How are the conditions changing over time? How do natural features and human activities affect the quality of streams and ground water, and where are those effects most pronounced? By combining information on water chemistry, physical characteristics, stream habitat, and aquatic life, the NAWQA Program aims to provide science-based insights for current and emerging water issues and priorities. NAWQA results can contribute to informed decisions that result in practical and effective water-resource management and strategies that protect and restore water quality.

Since 1991, the NAWQA Program has implemented interdisciplinary assessments in more than 50 of the Nation's most important river basins and aquifers, referred to as Study Units. (http://water.usgs.gov/nawqa/nawqamap.html). Collectively, these Study Units account for more than 60 percent of the overall water use and population served by public water supply, and are representative of the Nation's major hydrologic landscapes, priority ecological resources, and agricultural, urban, and natural sources of contamination.

Each assessment is guided by a nationally consistent study design and methods of sampling and analysis. The assessments thereby build local knowledge about water-quality issues and trends in a particular stream or aquifer while providing an understanding of how and why water quality varies regionally and nationally. The consistent, multi-scale approach helps to determine if certain types of water-quality issues are isolated or pervasive, and allows direct comparisons of how human activities and natural processes affect water quality and ecological health in the Nation's diverse geographic and environmental settings. Comprehensive assessments on pesticides, nutrients, volatile organic compounds, trace metals, and aquatic ecology are developed at the national scale through comparative analysis of the Study-Unit findings. (http://water.usgs.gov/nawqa/natsyn.html).



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# Abbreviations and Acronyms:

| | |
|---|---|
| µg/L | micrograms per liter |
| > | greater than |
| ≥ | greater than or equal to |
| Br | Bromide |
| $CaCl_2$ | Calcium chloride |
| $CHBr_2Cl$ | Dibromochloromethane |
| $CHBr_3$ | Tribromomethane (bromoform) |
| $CHBrCl_2$ | Bromodichloromethane |
| $CHCl_3$ | Trichloromethane (chloroform) |
| CFCs | Chlorofluorocarbons |
| Cl | Chloride |
| CWSs | Community water systems |
| HCFC-22 | Hydrochlorofluorocarbon-22 |
| MCL | Maximum Contaminant Level |
| HOBr | Hypobromous acid |
| HOCl | Hypochlorous acid |
| MTBE | Methyl-*tert* butyl ether |
| NaCl | Sodium chloride |
| NAWQA | National Water-Quality Assessment Program |
| PWSs | Public water systems |
| RCRA | Resource Conservation and Recovery Act |
| THMs | Trihalomethanes |
| TTHM | Total trihalomethane |
| TRI | Toxic Release Inventory |
| USEPA | U.S. Environmental Protection Agency |
| USGS | U.S. Geological Survey |
| VOCs | Volatile organic compounds |

# Sources and Occurrence of Chloroform and Other Trihalomethanes in Drinking-Water Supply Wells in the United States, 1986–2001

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## Abstract

Chloroform and three other trihalomethanes (THMs)—bromodichloromethane, dibromochloromethane, and bromoform—are disinfection by-products commonly produced during the chlorination of water and wastewater. Samples of untreated ground water from drinking-water supply wells (1,096 public and 2,400 domestic wells) were analyzed for THMs and other volatile organic compounds (VOCs) during 1986–2001, or compiled, as part of the U.S. Geological Survey's National Water-Quality Assessment Program. This report provides a summary of potential sources of THMs and of the occurrence and geographical distribution of THMs in samples from public and domestic wells. Evidence for an anthropogenic source of THMs and implications for future research also are presented.

Potential sources of THMs to both public and domestic wells include the discharge of chlorinated drinking water and wastewater that may be intentional or inadvertent. Intentional discharge includes the use of municipally supplied chlorinated water to irrigate lawns, golf courses, parks, gardens, and other areas; the use of septic systems; or the regulated discharge of chlorinated wastewater to surface waters or ground-water recharge facilities. Inadvertent discharge includes leakage of chlorinated water from swimming pools, spas, or distribution systems for drinking water or wastewater sewers. Statistical analyses indicate that population density, the percentage of urban land, and the number of Resource Conservation and Recovery Act hazardous-waste facilities near sampled wells are significantly associated with the probability of detection of chloroform, especially for public wells. Domestic wells may have several other sources of THMs, including the practice of well disinfection through shock chlorination, laundry wastewater containing bleach, and septic system effluent.

Chloroform was the most frequently detected VOC in samples from drinking-water supply wells (public and domestic wells) in the United States. Although chloroform was detected frequently in samples from public and domestic wells and the other THMs were detected in some samples, no concentrations in samples from either well type exceeded the U.S. Environmental Protection Agency's Maximum Contaminant Level of 80 micrograms per liter for total THMs. Chloroform was detected in public well samples almost twice as frequently (11 percent) as in domestic well samples (5 percent). The other three THMs also were detected more frequently in public well samples than in domestic well samples. This detection pattern may be attributed to public wells having a higher pumping capacity than domestic wells. The higher capacity wells create a larger capture zone that potentially intercepts more urban and other land uses and associated point and nonpoint sources of contamination than the smaller capacity domestic wells.

THM detection frequencies in domestic well samples show a pattern of decreasing frequency with increasing bromide content, that is in the order: chloroform > bromodichloromethane ≥ dibromochloromethane ≥ bromoform. This same pattern has been documented in studies of water chlorination, indicating that an important source of chloroform and other THMs in drinking-water supply wells may be the recycling of chlorinated water and wastewater. Mixtures of THMs commonly occur in public well samples, and the most frequently occurring are combinations of the brominated THMs. These THMs have limited industrial production, few natural sources, and small or no reported direct releases to the environment. Therefore, industrial, commercial, or natural sources are not likely sources of the brominated THMs in public and domestic well samples. The THM detection frequency pattern, the co-occurrence of brominated THMs, and other lines of evidence indicate that the recycling of water with a history of chlorination is an important source of these compounds in samples from drinking-water supply wells.

## Introduction

Chloroform—also known by its formal name as trichloromethane ($CHCl_3$)—was the most frequently detected volatile organic compound (VOC) in the Nation's ground water sampled by the U.S. Geological Survey's (USGS) National Water-Quality Assessment (NAWQA) Program (Squillace and others, 1999; Moran and others, 2002; Zogorski and others, 2006). Grady (2003) also reported chloroform as the most frequently detected VOC in a national study of both ground- and

surface-water sources of drinking water for community water systems (CWSs).

Chloroform and three other detected trihalomethanes (THMs)—bromodichloromethane, dibromochloromethane, and bromoform—are disinfection by-products commonly produced during the chlorination of water and wastewater. Strictly speaking, "trihalomethanes" applies to trihalogenated methanes containing any of the halogen atoms (chlorine, bromine, iodine, or fluorine), but this term will be used in this report to refer only to those containing either chlorine or bromine because these are the ones most commonly detected, or analyzed for, in chlorinated waters. The practice of treating large amounts of surface and ground water with chlorine for public health reasons began in the United States in Jersey City, New Jersey, in 1908, and since has become a standard procedure for CWSs across the Nation (Sawyer and McCarty, 1978). The primary purpose of chlorination of drinking water is to prevent the spread of waterborne diseases, especially fatal diseases such as cholera and typhoid. Although chlorination has been used to treat drinking water nationwide for almost 100 years, chloroform and other THMs were first identified by J.J. Rook in 1974 as being produced during the chlorination process (Rook, 1974).

The presence of chloroform in drinking water is a potential human-health concern. In 1986, the U.S. Environmental Protection Agency (USEPA) classified chloroform as a probable human carcinogen ("Group B2" carcinogen), based on evidence of its carcinogenicity in animals (U.S. Environmental Protection Agency, 2002). In 1998, the USEPA revised the carcinogenicity risk assessment of chloroform, stating that the compound is likely to be carcinogenic to humans by all routes of exposure at concentrations high enough to cause cytotoxicity and formation of regenerative nodules in susceptible tissues (U.S. Environmental Protection Agency, 2002). Bromodichloromethane and bromoform also are suspected human carcinogens, whereas insufficient data exist to assess the human carcinogenic potential of dichlorobromomethane (U.S. Environmental Protection Agency, 2004).

## Purpose and Scope

This report provides a summary of potential sources of THMs and of the occurrence and geographical distribution of THMs in samples collected or compiled by the USGS's NAWQA Program from drinking-water supply wells (public and domestic wells) during 1986–2001. Samples of untreated water (collected at the wellhead and before any treatment or blending) from 1,096 public and 2,400 domestic wells were collected nationally between 1986 and 2001 or compiled as part of the NAWQA Program. A detailed description of the public and domestic well data sets is documented in Zogorski and others (2006) and Moran and others (2006). Previously, Squillace and others (1999) reported the occurrence of THMs and other VOCs in untreated ambient ground water for samples collected during 1985–1995. Evidence of recycling of chlorinated water as a source of anthropogenic chloroform and other THMs also is presented in this report. Implications for future research also are described. A more detailed summary of chloroform sources (including information on industrial, degradation, and natural sources), formation of THMs as disinfection by-products, transport, fate, occurrence, and effects of THMs on human health and aquatic organisms is presented in Ivahnenko and Barbash (2004).

Public wells are privately or publicly owned and provide ground-water supply for public water systems (PWSs). In this report, the discussion of public wells refers to the quality of water captured by wells that provide drinking water to PWSs that serve at least 25 individuals. Domestic wells are defined as privately owned, self-supplied sources for household water use (Moran and others, 2002).

## Field Sampling and Laboratory Analytical Methods

NAWQA data-collection protocols and procedures for ground water (described in Koterba and others, 1995) were followed for the collection of samples for VOC analysis, as well as to characterize the water quality of the sampled aquifer. Laboratory analysis, including the description of the analytical methods and the associated quality-assurance/quality-control program, for the suite of 55 VOCs (including the four THMs) for the NAWQA Program are presented in Rose and Schroeder (1995) and Connor and others (1998). A summary of both the field and laboratory methods are described in Moran and others (2006).

In this report, all computations, including detection frequencies, were calculated at an assessment level of 0.2 microgram per liter (µg/L). The 0.2-µg/L assessment level represents the laboratory reporting value for the USGS prior to 1996 and for other data-contributing agencies. Because of improved field sampling protocols and analytical technology, the USGS laboratory reporting value was lowered after 1996; however, for consistency, concentrations of THMs in samples collected after 1996 were censored to 0.2 µg/L. The computation of total THMs in this report is the sum of all quantified concentrations of chloroform, bromodichloromethane, dibromochloromethane, and bromoform, censored to 0.2 µg/L.

## Acknowledgments

The author thanks all the PWS managers and private homeowners for allowing water samples to be collected from their wells. Through their cooperation, a nationwide data set of water-quality information has been generated, providing researchers with the information to assess the state of the Nation's aquifers and drinking-water supply wells at local, regional, and national levels. The author also thanks the many USGS field technicians, hydrologists, and laboratory personnel who have collected and analyzed the water samples during the first decade of the NAWQA Program.

# Potential Sources of Trihalomethanes to Drinking-Water Supply Wells

Chloroform and other THMs could originate from sources other than as by-products of water disinfection, and some of these industrial sources are documented on a nationwide basis in the USEPA's Toxic Release Inventory (TRI). In the United States, these direct releases to the environment have been reported annually and include air emissions, surface-water discharges, underground injections, and releases to land. According to the TRI, approximately 1.1 million pounds of chloroform were released in 2003 by these routes across the Nation (U.S. Environmental Protection Agency, 2005a). The paper industry contributed 52 percent of the total emissions (mostly as air emissions), the chemical industry contributed 28 percent (mostly as air emissions), the Resource Conservation and Recovery Act (RCRA) solvent recovery industry contributed 12 percent (nearly all as underground injections), and the food industry contributed 8 percent (all as air emissions) (U.S. Environmental Protection Agency, 2005a). According to the TRI, dichlorobromomethane and bromoform had total releases of 89 and 182 pounds, respectively, in 2003 (U.S. Environmental Protection Agency, 2005a). For both compounds, the chemical industry was the primary contributor, either as waste to landfills or as air emissions. Dibromochloromethane is not listed in the TRI compilations.

The major industrial use of chloroform is for the production of hydrochlorofluorocarbon-22 (HCFC-22), a refrigerant to replace the ozone-depleting chlorofluorocarbons (CFCs) in home air conditioners and large commercial freezers. In 1999, 98 percent of the industrially produced chloroform was used in the manufacture of HCFC-22. The remaining 2 percent was used for a variety of commercial purposes including reagents and extraction solvents (Budvari, 1989; Mannsville Chemical Products Corporation, 1999), fumigants, insecticides, and a precursor for dyes and pesticides (Lucius and others, 1992; Agency of Toxic Substances and Disease Registry, 1997). In 1998, the most recent year with production data, about 46 million gallons of chloroform were produced by industry.

Industrial use of the other THMs (bromodichloromethane, dibromochloromethane, and bromoform) has been as solvents for fats, waxes and resins, fire-extinguishing agents, and an intermediate in chemical synthesis. Currently (2005), dibromochloromethane has no commercial use (Agency of Toxic Substances and Disease Registry, 2005), and bromodichloromethane primarily is used only as a chemical intermediate for organic synthesis (Agency of Toxic Substances and Disease Registry, 1989). Because of the limited or non-existent use of both dibromochloromethane and bromodichloromethane, no information on industrial production volume for these compounds is available. However, total industrial production of bromoform in 1997, the most recent year with production data, was estimated to be as much as 45,700 gallons (Agency of Toxic Substances and Disease Registry, 2005).

An important source of anthropogenic chloroform detected in ground water and surface water appears to be chlorinated drinking water and wastewater that is discharged intentionally into the hydrologic system. This includes wastewater from the bleaching processes in pulp and paper mills, wastewater generated from the manufacturing and processing of chloroform-containing chemicals and materials, effluent from hazardous-waste disposal sites, and rinse water from domestic and industrial cleaning and laundry operations. Other such sources include regulated discharges of chlorinated water from wastewater treatment plants, cooling tower blowdown from electric power generating plants, and combined sewer overflows (Agency for Toxic Substances and Disease Registry, 1997). Most of these intentional discharges, however, are expected to represent only minor contributions to total chloroform releases to the hydrologic system, relative to the amount of chloroform contributed by inadvertent releases of chlorinated drinking water and wastewater (Agency for Toxic Substances and Disease Registry, 1997).

Chlorinated drinking water from PWSs may enter the subsurface through the irrigation of lawns, gardens, golf courses, athletic fields, and parks, as well as leaking swimming pools, spas, or sewers for treated wastewater. In addition, the maintenance of a chlorine residual is required in the distribution systems of PWSs and increases the likelihood that leakage from these systems also will lead to the contamination of ground water and surface water by chloroform and other disinfection by-products. Solley and others (1998) estimated that the loss of treated drinking water through leaking distribution pipes and other unknown routes may be as high as 15 percent in some systems. Overall, an estimated 0.4 million gallons of chloroform are generated per year through the chlorination of drinking water in the United States (McCulloch, 2003).

Although most homeowners who obtain their water from private wells do not chlorinate their water prior to use, chloroform was found to be the most frequently detected VOC in domestic well water. Potential sources of chloroform to domestic wells include (1) shock chlorination, which is a periodic disinfection process in which a dilute solution of bleach is added directly to a well to eliminate bacterial contamination, and (2) laundry wastewater containing bleach. In both cases, chloroform is produced through the haloform reaction of chlorine (contained in bleach) with organic matter.

Septic systems also may be an important source of chloroform to ground water. Previous studies have documented the presence of chloroform in septic-system effluent (DeWalle and others, 1985; Ayers Associates, 1993), and several States have identified septic systems as a major source of ground-water contamination, second only to underground storage tanks (U.S. Environmental Protection Agency, 2000). In addition, the U.S. Environmental Protection Agency (2000) has noted that improperly designed, maintained, or operated septic systems can result in ground-water contamination in the vicinity of the system, especially if the degradation of the organic matter is incomplete. The detection of chloroform or other THMs in a domestic well sample could be an indicator

of other possible health concerns (pathogenic microorganisms, for example) that may exist for the domestic well user (especially for those with untreated well water) whose well is in the vicinity of an improperly operating septic system.

Chloroform was originally considered of solely anthropogenic origins; however, several natural sources of chloroform have been identified. These include volcanic gases (Isidorov and others, 1990), marine algae (Gribble, 1994; Laturnus and others, 2002), and soil fungi (McCulloch, 2003). Although a natural substance, salt concentrations (NaCl and $CaCl_2$) have increased in the environment because of the use of these compounds for roadway deicing. A recent study (Princeton Geoscience, 2005) indicated that the abundance of chloride ions in the runoff of deicing meltwaters may be enhancing the natural chloroform formation process in nearby soils. Chloroform concentrations as high as 110 µg/L were determined in samples of shallow ground water adjacent to the paved surface. Although natural sources contribute approximately 90 percent of the total global calculated chloroform flux (McCulloch, 2003), evidence in the NAWQA-collected drinking-water well data is inconclusive about whether natural sources (including roadway deicing salts) contribute (and in what proportion) to chloroform in ground water. Marine algae also have been identified as natural sources for bromodichloromethane, dibromochloromethane, and bromoform (Gribble, 1994).

## Occurrence of Trihalomethanes in Drinking-Water Supply Wells

Of the 55 VOCs analyzed for by the NAWQA Program (Bender and others, 1999), chloroform was the most frequently detected VOC in samples from drinking-water supply wells. Chloroform was detected about twice as frequently in public well samples (11 percent) than in domestic well samples (5 percent), whereas other THMs occurred in public well samples 7 to 15 times more frequently than in domestic well samples (table 1). The other THMs (bromodichloromethane, dibromochloromethane, and bromoform) also were among the 10 most frequently detected VOCs in public well samples (fig. 1). Because the purpose of the NAWQA Program was to sample ambient, untreated ground water from different aquifers, the frequent and widespread occurrence of chloroform and the detection of other THMs in some samples was not expected.

The locations of the sampled wells and samples with total trihalomethane (TTHM) concentrations greater than 0.2 µg/L are shown in figure 2 for public wells and in figure 3 for domestic wells. Although chloroform and other THMs were detected frequently in public and domestic well samples, most TTHM concentrations were an order of magnitude lower than the USEPA Maximum Contaminant Level (MCL) of 80 µg/L. No individual THM concentrations in samples from either well type were greater than the USEPA's TTHM MCL of 80 µg/L (figs. 4 and 5). Median concentrations for individual THMs in samples with detections, from both public and domestic wells, ranged from 0.4 to 0.8 µg/L (figs. 4 and 5). Comparisons of THM concentrations to the USEPA's MCL are for informational purposes only, and do not indicate compliance with the drinking-water standard for water supplied by PWSs.

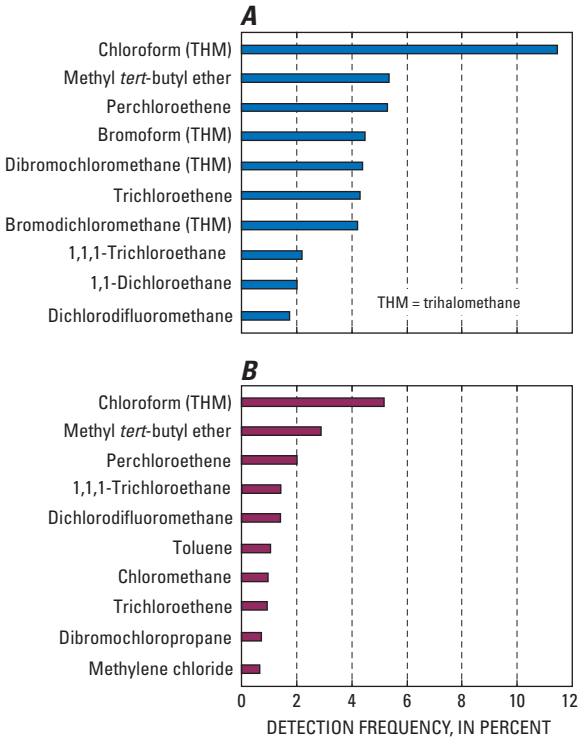


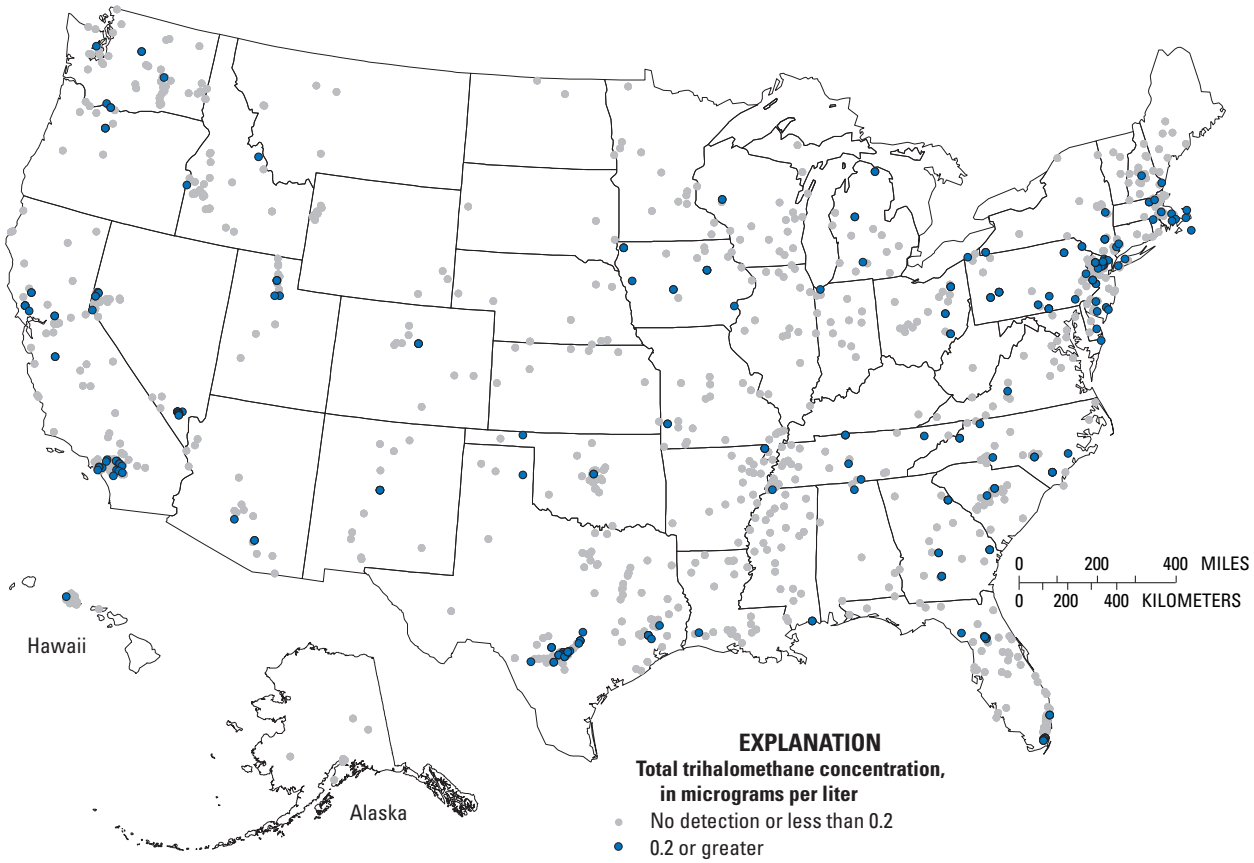

**Figure 1.** Detection frequencies of the 10 most frequently detected volatile organic compounds in (A) public well samples, and (B) domestic well samples at an assessment level of 0.2 microgram per liter.

## Evidence for an Anthropogenic Origin of Trihalomethanes in Drinking-Water Supply Wells

The greater frequency of THM detection in public well samples than in domestic well samples may be due to public wells having greater pumping capacities than domestic wells. These higher production capacities in public wells create a larger capture zone, greater drawdown, and faster movement of water from the top of the water table (including water containing surface-derived anthropogenic compounds that have migrated to the water table) to the well screen than the comparatively low production capacities of domestic wells (Bruce and Oelsner, 2001). The larger capture zone of the public wells also may include multiple land uses and point (as well as nonpoint) sources of VOCs (Stackelberg and others, 2000, 2001).

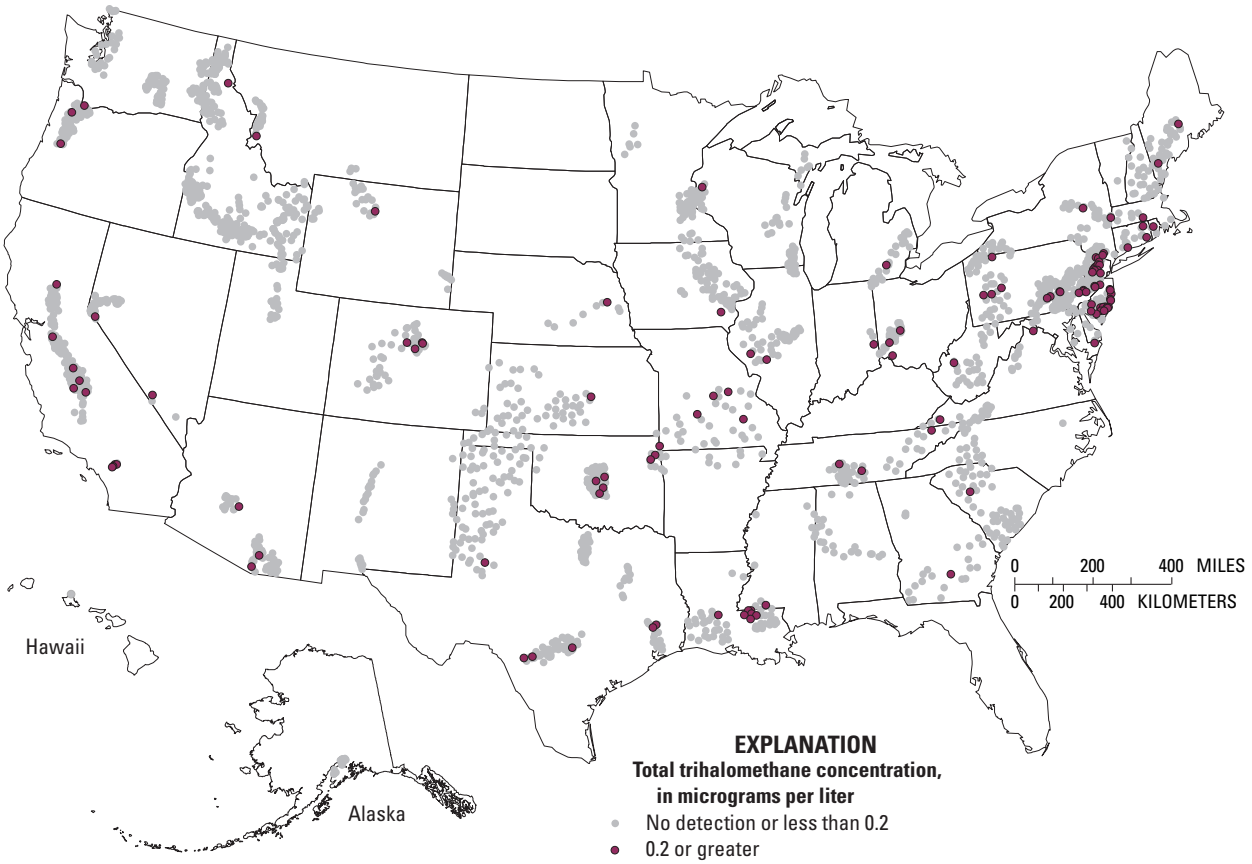
**Table 1.** Detection frequencies of trihalomethanes in samples from public and domestic wells at an assessment level of 0.2 microgram per liter.

| Compound name | Chemical formula | Number of samples | Number of detections | Detection frequency, in percent |
|---|---|---|---|---|
| | | Public wells | | |
| Chloroform | $CHCl_3$ | 1,092 | 125 | 11.4 |
| Bromodichloromethane | $CHBrCl_2$ | 1,095 | 46 | 4.2 |
| Dibromochloromethane | $CHBr_2Cl$ | 1,095 | 48 | 4.4 |
| Bromoform | $CHBr_3$ | 1,096 | 49 | 4.5 |
| | | Domestic wells | | |
| Chloroform | $CHCl_3$ | 2,400 | 124 | 5.2 |
| Bromodichloromethane | $CHBrCl_2$ | 2,400 | 14 | 0.6 |
| Dibromochloromethane | $CHBr_2Cl$ | 2,400 | 12 | 0.5 |
| Bromoform | $CHBr_3$ | 2,399 | 8 | 0.3 |




Base modified from U.S. Geological Survey digital data, 1:2,000,000, 1990
Albers Equal-Area Projection
North American Datum of 1983

**Figure 2.** Total trihalomethane concentrations of 0.2 microgram per liter or greater in public well samples.




Base modified from U.S. Geological Survey digital data, 1:2,000,000, 1990
Albers Equal-Area Projection
North American Datum of 1983

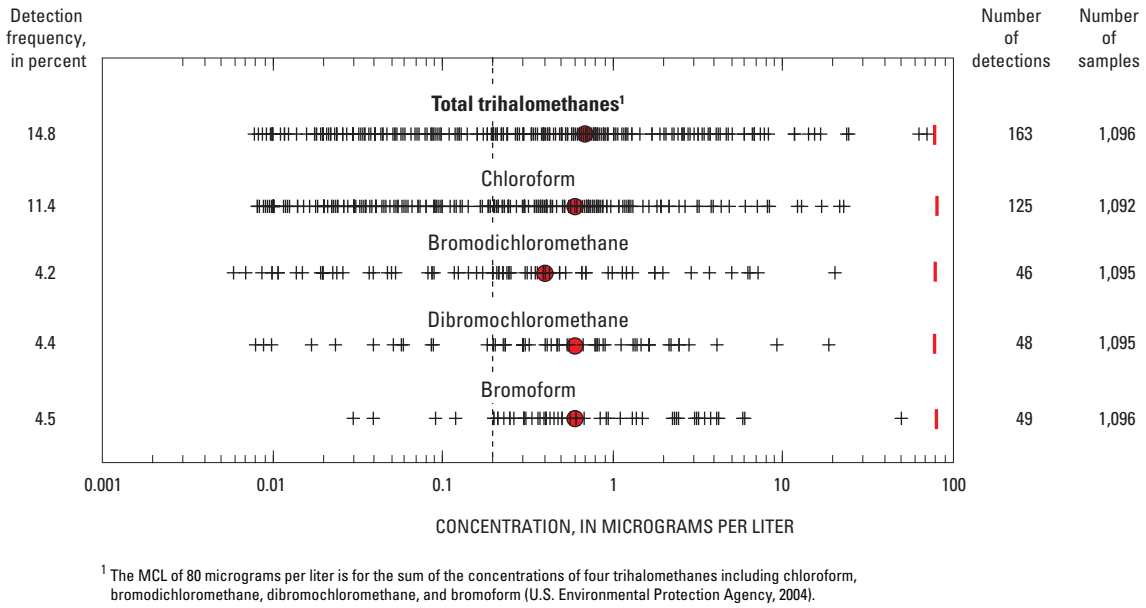
**Figure 3.** Total trihalomethane concentrations of 0.2 microgram per liter or greater in domestic well samples.

The greater detection frequency of chloroform and other THMs in public well samples than in domestic well samples also may be due to the larger likelihood of THM sources in the capture zone as indicated by the greater population density and greater amount of land classified as urban in proximity to public wells. Squillace and others (1999) reported that the probability of detecting one or more VOCs in ground water increased with increasing population density. Spatial analysis of 500-meter radius buffers around public and domestic wells using the 1990 Census data (U.S. Bureau of the Census, 1991) indicates that the buffers for public wells have an average population density about six times greater than buffers for domestic wells (table 2). In addition, a threefold greater percentage of the population was classified as living in urban areas near public wells than near domestic wells.

Chloroform and other THMs were detected in ground water beneath a broad range of land-use settings including not only urban, residential, and industrial locations, but also agricultural, forested, and other undeveloped areas. As might have been expected, among those studies that compared different land-use settings, detection frequencies of chloroform were higher beneath urban areas than beneath agricultural areas (fig. 6) (Stackelberg and others, 1997; Grady and Mullaney, 1998; Squillace and others, 1999). In addition, the percentage of urban land and RCRA hazardous-waste facilities near sampled wells have been shown to be factors statistically associated with the detections of chloroform in public and domestic well samples (Zogorski and others, 2006; Squillace and Moran, 2006).

**Table 2.** Population density and percent of population classified as living in urban land use in 500-meter radius buffer around public and domestic wells.

| Type of wells | Average population density (persons per square mile) | Urban population as percentage of total population |
|---|---|---|
| Public wells | 423 | 41 |
| Domestic wells | 73 | 14 |




[1] The MCL of 80 micrograms per liter is for the sum of the concentrations of four trihalomethanes including chloroform, bromodichloromethane, dibromochloromethane, and bromoform (U.S. Environmental Protection Agency, 2004).

**EXPLANATION**

+ **VOC concentration for a sample**—Results from several analytical methods, with differing sensitivity for a specific VOC with time, as well as between VOCs, are shown in the bar concentration chart. Multiple samples of equal concentration will appear as a single symbol

● **Median concentration**

14.8 **Detection frequency, in percent, at an assessment level of 0.2 microgram per liter**

163 **Number of detections at an assessment level of 0.2 microgram per liter**

1,096 **Number of samples**

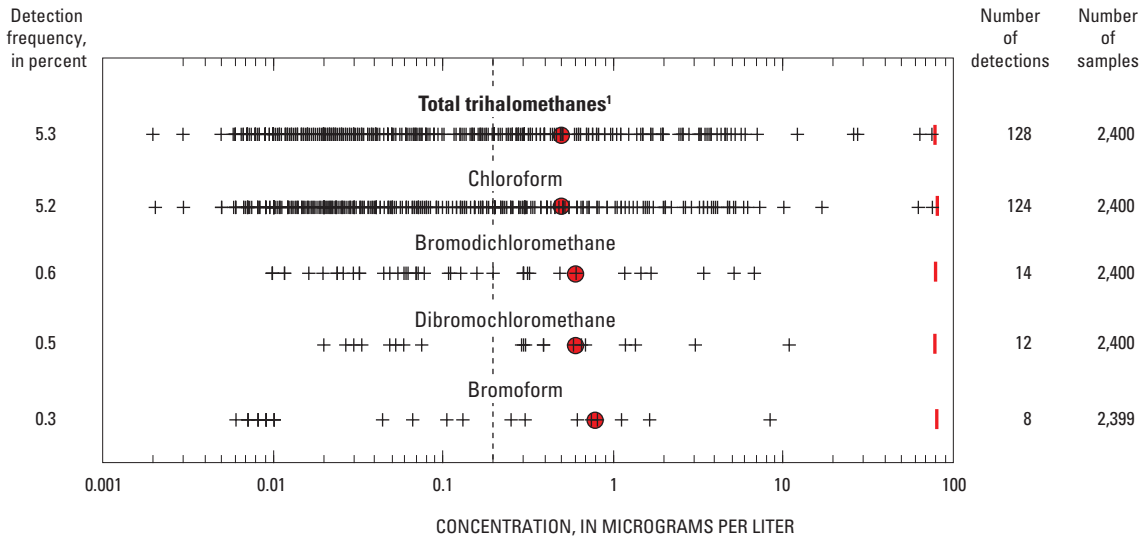
| **U.S. Environmental Protection Agency Maximum Contaminant Level (MCL) for total trihalomethanes**

------ **Assessment level of 0.2 microgram per liter**

**Figure 4.** Concentrations of trihalomethanes in public well samples.

When the bromide ion is present during chlorination, either as a naturally occurring substance or as a commonly occurring contaminant in the chlorine used for disinfection (U.S. Environmental Protection Agency, 2005b), the haloform reaction generally produces one or more of the brominated THMs in addition to chloroform (Boyce and Hornig, 1983). This occurs even if bromide concentrations are several orders of magnitude lower than chlorine concentrations on a molar basis (Luong and others, 1982). Formation of brominated THMs occurs under such conditions because (1) free chlorine oxidizes bromide to free bromine ($HOCl + Br^- \rightarrow HOBr + Cl^-$) much more rapidly than it reacts with dissolved organic matter (Luong and others, 1982), and (2) free bromine is a more powerful halogenating agent than free chlorine (Rook, 1974).

At low molar ratios of bromide ion to free chlorine (0.03 or less), the yields of the four principal THMs generated during chlorination of water decrease with increasing bromination of the product in the following order: $CHCl_3 > CHBrCl_2 \geq CHBr_2Cl \geq CHBr_3$ (Luong and others, 1982; Summers and others, 1993). This trend also was evident among the concentrations of the THMs formed during the study of the THM formation potential of agricultural drainage waters from the Sacramento-San Joaquin Delta, California (Fujii and others, 1998). Results from studies where more than one THM has been detected in the hydrologic system commonly show the same trend among the detection frequencies for these compounds (Rook, 1974; Westrick and others, 1984; Voelker, 1989; Kolpin and Thurman, 1995; Stackelberg and others, 1997; Reiser and O'Brien, 1998; Lungren and Lopes, 1999; Squillace and others, 1999; Inkpen and others, 2000; Thiros, 2000; Grady and Casey, 2001; Moran and others, 2002). Such observations indicate that the presence of brominated THMs might be used as a valuable criterion for distinguishing chlorination-treated waters from other potential sources of chloroform in the hydrologic system—especially if the concentrations among the four principal THMs (or detection




[1] The MCL of 80 micrograms per liter is for the sum of the concentrations of four trihalomethanes including chloroform, bromodichloromethane, dibromochloromethane, and bromoform (U.S. Environmental Protection Agency, 2004).

**EXPLANATION**

| | |
|---|---|
| + | **VOC concentration for a sample**—Results from several analytical methods, with differing sensitivity for a specific VOC with time, as well as between VOCs, are shown in the bar concentration chart. Multiple samples of equal concentration will appear as a single symbol |
| ● | **Median concentration** |
| 5.3 | **Detection frequency, in percent, at an assessment level of 0.2 microgram per liter** |
| 128 | **Number of detections at an assessment level of 0.2 microgram per liter** |
| 2,400 | **Number of samples** |
| \| | **U.S. Environmental Protection Agency Maximum Contaminant Level (MCL) for total trihalomethanes** |
| - - - - - - | **Assessment level of 0.2 microgram per liter** |

**Figure 5.** Concentrations of trihalomethanes in domestic well samples.

frequencies, if based on the same detection limit for all four compounds) have the trend noted above.

Detection frequencies of THMs in samples from domestic wells show the trend of decreasing detection frequency with increasing bromide, that is, in the following order: $CHCl_3 > CHBrCl_2 \geq CHBr_2Cl \geq CHBr_3$ (table 1), indicating that chlorinated water may be the source. This pattern is less evident for public well samples; however, mixtures of two of more THMs occurred in 5.4 percent of public well samples (table 3), or about 37 percent of the samples that had one or more THM present. Of these mixtures, the most frequently occurring were combinations of the brominated THMs (table 4). In addition, the percentage of public well samples with detections of one or more THMs decreases with the number of THMs detected (table 3), whereas this pattern is not as evident for domestic well samples. In general, the brominated THMs are more predominant in public well samples than in domestic well samples, indicating that the capture of chlorinated water may be the primary source of THMs in public well samples; however, the detection of all four of the THMs in domestic well samples also indicates an anthropogenic source for these wells.

The brominated THMs have limited industrial production, few natural sources, and small or no reported direct releases to the environment, as noted previously. Therefore, industrial, commercial, or natural sources are not likely sources of the brominated THMs to public and domestic wells. Furthermore, the national distribution (figs. 2 and 3) and occurrence of chloroform and the brominated THMs in both public and domestic well samples and the occurrence pattern ($CHCl_3 > CHBrCl_2 \geq CHBr_2Cl \geq CHBr_3$) for domestic well samples indicates that the source of the compounds may be the result of the nationwide practice of water chlorination and use of bleach in households.

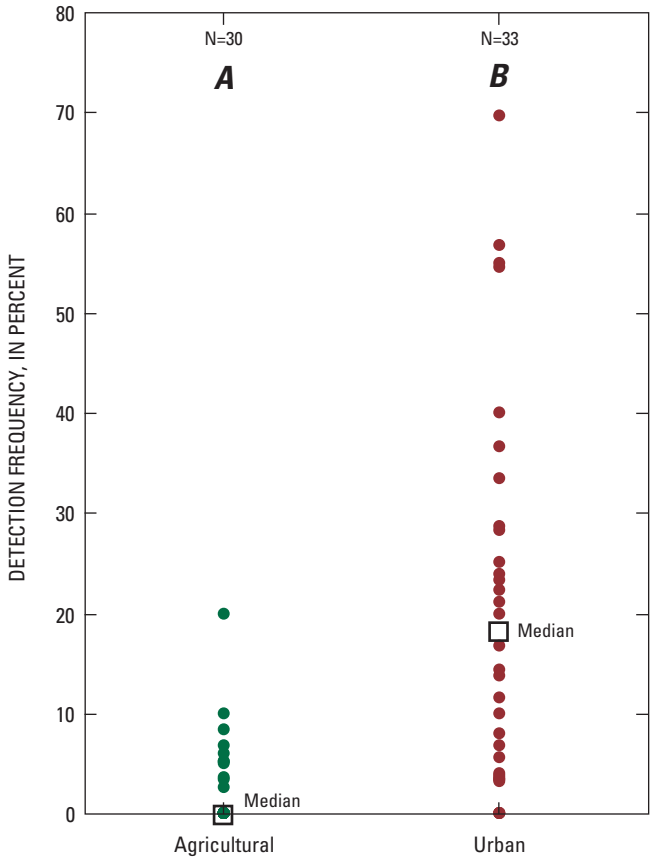



**Figure 6.** Detection frequencies of chloroform at 0.2 microgram per liter or greater in agricultural and urban land-use areas (The different letters (A, B) indicate statistical difference at the 0.05 significance level between the land-use areas; each dot represents a separate land-use study.

**Table 3.** Number of trihalomethanes detected at an assessment level of 0.2 microgram per liter and the percentage of samples with detections in samples from public and domestic wells, 1986-2001.

| Category | Number of trihalomethanes detected | | | | | |
|---|---|---|---|---|---|---|
| | None | One | Two | Three | Four | Two or more |
| Public Wells | | | | | | |
| Number of samples | 935 | 103 | 25 | 17 | 16 | 58 |
| Percentage of samples with detections | 85.3 | 9.4 | 2.3 | 1.6 | 1.5 | 5.4 |
| Domestic Wells | | | | | | |
| Number of samples | 2,274 | 111 | 4 | 5 | 6 | 15 |
| Percentage of samples with detections | 94.8 | 4.6 | .2 | .2 | .2 | .6 |

**Table 4.** Most frequently occurring two-compound mixtures in public well samples at an assessment level of 0.2 microgram per liter.

[VOC, volatile organic compound; MTBE, methyl-*tert* butyl ether]

| Rank | Mixture | | Number of detections | Detection frequency, in percent |
|---|---|---|---|---|
| | Trihalomethane | Second VOC | | |
| 1 | Bromodichloromethane | Dibromochloromethane | 37 | 3.4 |
| 2 | Chloroform | Bromodichloromethane | 35 | 3.2 |
| 3 | Dibromochloromethane | Bromoform | 32 | 2.9 |
| 4 | Chloroform | Dibromochloromethane | 27 | 2.5 |
| 5 | Bromodichloromethane | Bromoform | 22 | 2.0 |
| 6 | Chloroform | Bromoform | 19 | 1.7 |
| 7 | Chloroform | Trichloroethene | 17 | 1.6 |
| 8 | Chloroform | Perchloroethene | 15 | 1.4 |
| 9 | Chloroform | 1,1,1-Trichloroethane | 14 | 1.3 |
| 10 | Chloroform | MTBE | 13 | 1.2 |

## Implications for Future Research

The widespread occurrence of chloroform in samples from drinking-water supply wells may be especially important with regards to the presence of other disinfection by-products, for example, haloacetic acids, which have a USEPA MCL of 60 µg/L for the total concentration of five compounds (U.S. Environmental Protection Agency, 2001). Studies to assess the extent of the occurrence, transport pathways, and concentration ranges of haloacetic acids and other disinfection by-products are needed because little national, regional, or aquifer-scale data exist for these compounds, especially for domestic wells. Other areas of possible future research include more detailed information regarding the spatial distributions and temporal variability of anthropogenic sources of THMs, especially chlorinated drinking water and wastewater derived from leaking water distribution lines and wastewater sewers, as well as chlorinated water used to irrigate lawns, golf courses, parks, gardens, and other urban and residential settings.

## Summary

Chloroform and three other trihalomethanes (THMs)—bromodichloromethane, dibromochloromethane, and bromoform—are disinfection by-products commonly produced during the chlorination of water and wastewater. As part of the U.S. Geological Survey's National Water-Quality Assessment (NAWQA) Program, samples of untreated ground water from drinking-water supply wells (1,096 public and 2,400 domestic wells) were analyzed for THMs and other volatile organic compounds (VOCs) during 1986–2001, or compiled from other studies. This report provides a summary of potential sources of THMs and of the occurrence and geographical distribution of THMs in these samples and presents evidence for an anthropogenic source of chloroform and other THMs. Implications for future research also are presented.

Potential sources of THMs to both public and domestic wells include the use and discharge of chlorinated drinking water and wastewater that may be intentional. These potential sources include the use of municipally supplied chlorinated water to irrigate lawns, golf courses, parks, gardens, and other areas; the use of septic systems; or the regulated discharge of chlorinated wastewater to surface waters or ground-water recharge facilities. Other potential sources of THMs to public and domestic wells also may be inadvertent including leakage of chlorinated water from swimming pools, spas, or distribution systems for drinking water or wastewater sewers.

Although most water from domestic wells is not chlorinated prior to use, chloroform was found to be the most frequently detected VOC in domestic well samples. Several potential additional sources of chloroform include shock chlorination, in which a dilute solution of bleach is added directly to the well to eliminate bacterial contamination, and laundry wastewater containing bleach. In both cases, chloroform is produced through the haloform reaction of chlorine (contained in bleach) with organic matter. Also, chloroform has

been documented in septic-system effluent, and improperly designed, maintained, or operated septic systems can result in ground-water contamination in the vicinity of the system. The detection of chloroform or other THMs in a domestic well sample could be an indicator of other possible health concerns (pathogenic microorganisms, for example) that may exist for the domestic well user (especially for those with untreated well water) whose well is in the vicinity of an improperly operating septic system.

Chloroform also was the most frequently detected VOC in public well samples collected or compiled by the U.S. Geological Survey's NAWQA Program. The other THMs—bromodichloromethane, dibromochloromethane, and bromoform—were among the 10 most frequently detected VOCs in the NAWQA data sets for public well samples. The widespread occurrence and frequency of THMs in public and domestic well samples, however, was not expected, as the purpose of the NAWQA Program was to sample ambient, untreated ground water from a large number of aquifers across the Nation. Although chloroform was detected frequently in public and domestic well samples and other THMs were present in some samples, no total THM concentrations in samples from either well type exceeded the U.S. Environmental Protection Agency's Maximum Contaminant Level of 80 micrograms per liter (μg/L).

Chloroform was detected about twice as frequently in public well samples than in domestic well samples, which may be due to the generally higher capacity public wells generating a larger capture zone in land-use areas classified as urban. The larger capture zone for public wells could incorporate multiple land uses and a greater number of point and nonpoint contamination sources than the smaller capture zone of domestic wells. Chloroform was detected in ground water in a broad range of land-use settings; however, the detection frequencies of chloroform were higher beneath urban areas than beneath agricultural areas. Statistical analyses indicate that population density, percentage of urban land, and the number of Resource Conservation and Recovery Act hazardous-waste facilities near the sampled wells have a significant association with the detection of chloroform.

THM detection frequencies for public and domestic well samples at an assessment level of 0.2 μg/L showed a general pattern of decreasing frequency with increasing bromide content, that is chloroform > bromodichloromethane ≥ dibromochloromethane ≥ bromoform. This pattern is more evident for the domestic well samples than for the public well samples. Because this same detection frequency pattern of chloroform > bromodichloromethane ≥ dibromochloromethane ≥ bromoform has been reported previously for chlorinated waters, it might be used as a criterion for distinguishing THMs resulting from water with a history of chlorination from other potential sources of chloroform and other THMs in the hydrologic system.

Chloroform in drinking-water supply wells could originate from many sources. However, the observation that chloroform commonly occurred in mixtures with the other THMs, especially in public well samples, and that the detection frequency of the THMs was in the following order: chloroform > bromodichloromethane ≥ dibromochloromethane ≥ bromoform in many of the public and domestic well samples, indicate that chlorinated drinking water and/or wastewater are important sources of THMs in samples from supply wells. Additionally, the brominated THMs have limited industrial production, few natural sources, and small or no reported direct releases to the environment, and are not likely sources of the brominated THMs in public and domestic well samples.

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