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# Pharmaceutical Compounds in Merrimack River Water Used for Public Supply, Lowell, Massachusetts, 2008–09

Scientific Investigations Report 2011–5192

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

**Cover.** The Pawtucket Falls Dam at Lowell, Massachusetts (Photograph taken December 17, 2010).

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By Andrew J. Massey and Marcus C. Waldron

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**U.S. Department of the Interior**
KEN SALAZAR, Secretary

**U.S. Geological Survey**
Marcia K. McNutt, Director

U.S. Geological Survey, Reston, Virginia: 2011

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# Conversion Factors, Datum, and Acronyms

Inch/Pound to SI

| Multiply | By | To obtain |
|---|---|---|
| | Length | |
| inch (in.) | 2.54 | centimeter (cm) |
| foot (ft) | 0.3048 | meter (m) |
| mile (mi) | 1.609 | kilometer (km) |
| | Area | |
| square mile ($mi^2$) | 259.0 | hectare (ha) |
| square mile ($mi^2$) | 2.590 | square kilometer ($km^2$) |
| | Volume | |
| gallon (gal) | 3.785 | Liter (L) |
| million gallons (Mgal) | 3,785 | cubic meter ($m^3$) |
| cubic foot ($ft^3$) | 0.02832 | cubic meter ($m^3$) |
| | Flow rate | |
| cubic foot per second ($ft^3/s$) | 0.02832 | cubic meter per second ($m^3/s$) |
| million gallons per day (Mgal/d) | 0.04381 | cubic meter per second ($m^3/s$) |
| | Mass | |
| ounce, avoirdupois (oz) | 28.35 | gram (g) |

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$°F=(1.8×°C)+32$$

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows:

$$°C=(°F–32)/1.8$$

Vertical coordinate information is referenced to the North American Vertical Datum of 1988 (NAVD 88).

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Altitude, as used in this report, refers to distance above the vertical datum.

# Acronyms

| | |
|---|---|
| CFU | colony-forming units |
| CSO | combined sewer overflow |
| CSS | combined sewer system |
| CWA | Clean Water Act |
| DWTP | drining-water treatment plant |
| LRWU | Lowell Regional Water Utility |
| MassDEP | Massachusetts Department of Environmental Conservation |
| NAWQA | National Water-Quality Assessment |
| NPDES | National Pollution Discharge Elimination System |
| OWC | organic wastewater contaminants |
| QA | quality assurance |
| QC | quality control |
| RL | reporting level |
| SWQA | Source-Water Quality Assessment |
| USGS | U.S. Geological Survey |
| WWTP | wastewater-treatment plant |

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# Pharmaceutical Compounds in Merrimack River Water Used for Public Supply, Lowell, Massachusetts, 2008–09

By Andrew J. Massey and Marcus C. Waldron

## Abstract

This report presents results of a study conducted by the U.S. Geological Survey (USGS), in cooperation with the Massachusetts Department of Environmental Protection, to determine the occurrence of 14 commonly used human-health pharmaceutical compounds and fecal-indicator bacteria in Merrimack River water used as a drinking-water source by 135,000 residents in eastern Massachusetts. The study was designed to complement the USGS National Water-Quality Assessment Program's Source Water-Quality Assessment, which identifies patterns of occurrence of 280 primarily unregulated organic wastewater contaminants in source water used by community water systems and determines whether these patterns also occur in treated drinking water prior to distribution. The study involved periodic collection and analysis of raw Merrimack River water and treated drinking water over the course of 1 year. Water samples were collected periodically without regard to flow regime or antecedent weather conditions at the Lowell Regional Water Utility's Merrimack River intake upstream from Lowell, Mass. The same parcel of water was then sampled as finished water following treatment.

Despite the presence of many potential sources of contamination in the drinking-water source area, only 2 of the 14 pharmaceutical analytes were detected at reportable concentrations in the source-water samples, and these occurred in only one set of periodic samples. Acetaminophen, a nonprescription analgesic, and caffeine were detected in the September source-water samples at concentrations of 0.084 and 0.068 micrograms per liter, respectively. Three other compounds—carbamazepine, an antiepileptic; cotinine, a metabolite of nicotine; and diphenhydramine, a nonprescription antihistamine—were detected in source-water samples, but at concentrations too low to be reliably quantified. None of the 14 pharmaceuticals was found in the finished water at a reportable concentration, defined as two times the long-term detection limit used by the analytical laboratory.

In addition to the pharmaceutical analyses, measurements of fecal-indicator bacteria (*Escherichia coli*) concentrations and several physical characteristics were made on all source-water samples. Values for these constituents were consistently within State standards. It is possible that the monthly sampling schedule missed hydrologic events that would have transported greater concentrations of sewage contaminants to the sampling site, or that the large flow volume of the river at the study site effectively diluted the contaminant signal, but it is also likely that recent efforts to separate stormwater- and wastewater-discharge systems in the reaches upstream from the Lowell Regional Water Utility have greatly reduced the potential for sewage contamination at the intake.

## Introduction

In the United States and elsewhere, reports of the occurrence and distribution of organic wastewater contaminants (OWCs), including antibiotics, pharmaceuticals, natural and synthetic hormones, and industrial and household products (for example, pesticides, flame retardants, detergent metabolites, antioxidants, and disinfectants) in drinking-water supplies have become commonplace (Barnes and others, 2008; Focazio and others, 2008). A U.S. Geological Survey (USGS) national survey reported the presence of at least one of these compounds in 80 percent of 139 streams sampled in 1999–2000 (Kolpin and others, 2002). In general, concentrations of OWCs detected in the USGS survey were less than 1 part per billion, but 75 percent of streams had more than 1 compound, and 34 percent had 10 or more. The health effects of exposure to low concentrations and complex mixtures of these compounds are largely unknown. Human or ecological health standards or criteria have been established for only a very small number of OWCs.

In response to the need for more detailed information on the occurrence and distribution of these compounds in the Nation's drinking-water supplies, the USGS initiated a Source Water-Quality Assessment (SWQA) as part of its National Water-Quality Assessment (NAWQA) Program. The goal of the SWQA is to characterize the quality of selected rivers and aquifers used as a source of supply to community water systems throughout the United States (Delzer and Hamilton, 2007). Surface-water assessments consist of sampling at the intakes of surface-water supplies and resampling of the same water after passage through the drinking-water treatment process. Sampling is conducted monthly to account for seasonal

variability, and additional source and finished-water sample pairs are collected during hydrologic events when conditions are changing rapidly and contaminant concentrations are expected to be elevated. For groundwater assessments, samples from high-production wells supplying community water systems and the associated finished water are collected as described in Carter and others (2010). Source-water samples are analyzed for the fecal-indicator bacterium *Escherichia coli* (*E. coli*), and all samples are analyzed for 280 OWCs, including (1) disinfection byproducts; (2) fumigant-related compounds; (3) fungicides; (4) gasoline hydrocarbons, oxygenates, and oxygenate degradates; (5) herbicides and herbicide degradates; (6) insecticides and insecticide degradates; (7) manufacturing additives; (8) organic synthesis compounds; (9) pavement- and combustion-derived compounds; (10) personal-care and domestic-use products; (11) plant- or animal-derived biochemicals; (12) refrigerants and propellants; and (13) solvents.

This report describes the occurrence of 14 commonly used prescription and nonprescription pharmaceutical compounds in source water from the Merrimack River in northeastern Massachusetts and in the same water after treatment at the Lowell Regional Water Utility (LRWU). The LRWU community water system provides treated drinking water for more than 135,000 residents, businesses and other customers in the communities of Lowell, Dracut, Tyngsborough, and Chelmsford, Mass., and typically produces about 4.7 billion gallons of drinking water annually (The City of Lowell, 2010). The Merrimack River and the LRWU were sampled in 2008–09 as part of the USGS SWQA. However, because pharmaceuticals were not included in the original SWQA analyte list, they were added to the Merrimack River assessment through a cooperative agreement between the USGS and the Massachusetts Department of Environmental Protection. Table 1 provides a list of the compounds together with their Chemical Abstracts Service (CAS) Registry numbers and primary uses.

Water samples were collected periodically from April 2008 through June 2009 and included 12 source-water samples paired with 12 finished-water samples, plus several quality-assurance (QA) samples. Measurements of several physical characteristics were recorded for the water samples, and additional source-water samples were collected for determination of concentrations of *E. coli*. Analytical results for the 280 SWQA target-analyte-list compounds are published separately in Carter and others (2010).

**Table 1.** Human-health pharmaceutical compounds measured in raw and finished water from the Merrimack River at the Lowell Regional Water Utility, Massachusetts, 2008–09.

| Compound name | Chemical Abstracts Service Registry number | Primary use |
|---|---|---|
| 1,7-Dimethylxanthine | 611-59-6 | Caffeine metabolite |
| Acetaminophen | 103-90-2 | Antipyretic |
| Albuterol (Salbutamol) | 18559-94-9 | Antiasthmatic |
| Caffeine | 58-08-2 | Stimulant |
| Carbamazepine | 298-46-4 | Antiepileptic |
| Codeine | 76-57-3 | Analgesic |
| Cotinine | 486-56-6 | Nicotine metabolite |
| Dehydronifedipine | 67035-22-7 | Antianginal |
| Diltiazem | 42399-41-7 | Antihypertensive |
| Diphenhydramine | 147-24-0 | Antihistamine |
| Sulfamethoxazole | 723-46-6 | Antibiotic |
| Thiabendazole | 148-79-8 | Antifungal, antihelmintic |
| Trimethoprim | 738-70-5 | Antibiotic |
| Warfarin | 81-81-2 | Anticoagulant |

## Hydrologic Setting of the Merrimack River Drinking-Water Source

The Merrimack River watershed is the fourth largest in New England with a drainage area of 5,010 square miles ($mi^2$) about one quarter of this area is in Massachusetts (fig. 1). The headwaters of the Merrimack River originate more than 175 miles (mi) from the mouth in alpine and subalpine streams on the western and southern aspects of the White Mountain section of the New England physiographic province in central New Hampshire. The main stem of the Merrimack River begins south of the White Mountain National Forest at the confluence of the Pemigewasset River and the Winnipesaukee River. From this confluence, the river flows 115 mi through the uplands of south-central and southern New Hampshire into Massachusetts. In Massachusetts the river arcs to the northeast, flowing roughly parallel to the New Hampshire border and then draining to the Atlantic Ocean at Newburyport, Mass. (fig. 1).

The main sources of river flow are precipitation and snowmelt totaling about 45 inches (in.) per year; greater amounts (up to 70 in.) of annual precipitation fall at headwater altitudes. Groundwater is also a very important component of the Merrimack River flow. Annual precipitation recharges groundwater, which, in turn, contributes up to 60 percent of annual river flow (New England River Basins Commission, 1978) through permeable sand and gravel aquifers along the river and its tributaries. The Merrimack River watershed is geographically diverse and ranges from steep granite and gneiss alpine summits in the north to the relatively flat coastal basin near the Atlantic coast in northeastern Massachusetts. The final 22-mi reach of the Merrimack River, downstream from Haverhill, Mass., is tidally influenced. Between the White Mountains and Atlantic Ocean, the Merrimack River Valley widens, and the river and its tributaries provide a level flood plain, which is used for varied agricultural production.

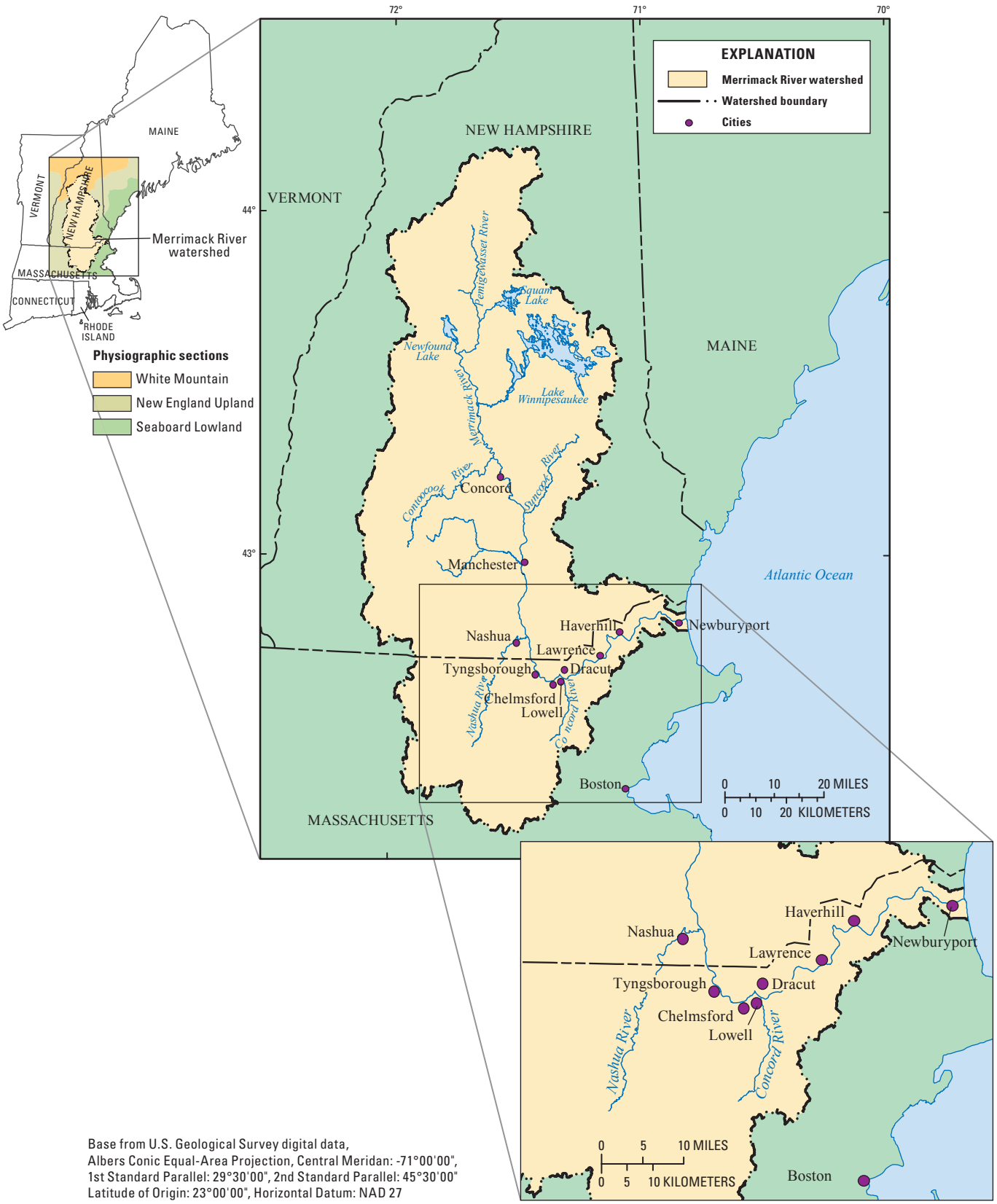


**Figure 1.** Map showing the Merrimack River watershed in New Hampshire and Massachusetts and the location of the study area on the Merrimack River near Lowell, Massachusetts.

## Sources of Contamination

Land use in the Merrimack River watershed ranges from undeveloped State lands—National Forest and wetlands—to farmland and agriculture, and from small rural towns to relatively large industrial cities, including Manchester and Nashua, N.H., and Lowell and Lawrence, Mass. Most population centers developed along rivers that powered early textile and industrial mills. These modern cities that have developed from the early mill towns require centralized drinking-water and wastewater-treatment systems to maintain sanitary conditions and safe, reliable drinking water. The wastewater generally discharges to the river at local outfalls. In 1990, about half (48 percent) of New Hampshire's 1.1 million residents were connected to a municipal sewer system (Medalie, 1996). In rural unsewered areas, onsite septic systems are typically used for the disposal of residential wastewater.

The Merrimack River receives treated sewage effluent in the form of point-source return flows from 46 wastewater-treatment plants (WWTP) permitted to discharge to the main stem and tributaries, including the three largest municipal WWTP in New Hampshire. Of the total return flows from municipal facilities in New Hampshire, 50 million gallons per day (Mgal/d), or 60 percent, discharges to surface water in the Merrimack River Basin. There are also potential sources of contamination from municipal wastewater disposal into the Massachusetts reach of the Merrimack River. Municipal WWTPs in the Nashua River Basin contribute an additional 22.5 Mgal/d of treated return flow upstream from the LRWU drinking-water intake. Another 57.3 Mgal/d of wastewater returns to streams in the Merrimack River Basin downstream from the LRWU intake (Medalie, 1996).

Point-source wastewater-effluent outfalls present a considerable potential source of contamination to the Merrimack River drinking-water supply; however, these point sources are highly regulated through the National Pollution Discharge Elimination System (NPDES), which includes effluent standards and requires that wastewater-treatment technologies be developed to protect the aquatic environment and minimize risks to human health. These standards have been particularly important in reducing bacteria and nutrients that once resulted in unsanitary conditions and decreased ecological health in the receiving waters near large population centers. The treatment process, however, was not designed to remove the entire range of anthropogenic materials in the waste stream. Additionally, many anthropogenic chemicals in the environment remain unregulated and, in general, their collective toxicity is poorly understood.

The NPDES, authorized by the Clean Water Act (CWA) in 1972 (http://cfpub.epa.gov/npdes/cwa.cfm?program_id=6) imposed statutory requirements including industry-wide technology-based standards for permitted discharges of treated effluent to receiving surface waters. Prior to the establishment of the NPDES, it was common for raw sewage to be dumped directly into the environment, and many sewage plants performed only primary treatment of wastewater prior to release. Primary treatment is the process of allowing solids to settle out of the waste stream prior to discharge, but the method is usually only about 30 to 60 percent effective at removing suspended solids. Secondary treatment follows primary treatment and is designed to remove organic matter through biological processes that result in the settling and separation of organic-rich sludge from the clearer supernatant. Modern wastewater-treatment technologies also typically include a final disinfection process (irradiation by ultraviolet light or chlorination, for example) to kill any remaining bacteria or other pathogens. Secondary treatment at the Nashua, N.H., WWTP has been online since October 1989 (Mario Leclerc, Superintendent, Nashua Wastewater Treatment Facility, written commun., 2010). Similar technologies are used at the Manchester, N.H., WWTP, from which the average daily discharge of treated effluent is about 25 Mgal/d.

Combined sewer systems (CSS), in which sewage and stormwater are conveyed to a treatment plant through a single pipe, may also affect water quality in the Merrimack Basin. CSSs may become overwhelmed during heavy rainfall events, causing a combined sewer overflow (CSO). CSOs may convey untreated domestic, commercial, and industrial wastes along with stormwater runoff into receiving waters such as the Merrimack River main stem. Major efforts have been underway since the 1990s to address CSSs in New Hampshire. Since 2003, four of the six cities in New Hampshire with CSSs have successfully separated their stormwater and wastewater systems. Projects in Nashua and Manchester have successfully eliminated some CSSs, and more projects are planned or proposed to correct the remaining CSSs, where CSOs still occur during heavy precipitation (Mucciarone, 2010). In Manchester, N.H., heavy rainfall causes approximately 44 CSOs in an average year (Fredrick J. McNeill, city of Manchester, written commun., 2010). On an annual basis, CSOs may contribute smaller amounts of fecal coliform and *E. coli* bacteria to the river system than other nonpoint sources to the Merrimack River; simulation results attribute 19 and 8 percent, respectively, of the annual loads of these contaminants to CSOs (Camp Dresser & McKee, Inc., 2004). On a daily or weekly basis, however, CSOs may be the main source of these contaminants to downstream sections of the river. In fact, procedures requiring WWTP operators to inform downstream water users of CSOs or other unplanned discharge events are in place so that adjustments may be made to drinking-water treatment processes in a timely manner. Recreational activities involving human contact with affected surface waters are strongly discouraged during CSO events.

Another potential concern regarding wastewater discharged to receiving waters is the persistence of certain pharmaceutical compounds in the environment after disposal. Recent research has shown that many pharmaceutical compounds and other OWCs pass unaffected through wastewater treatment and may persist in surface water and groundwater at distance from the sources (Barnes and others, 2008; Kolpin and others, 2002). In rural areas where household wastewater is treated with onsite septic systems, groundwater flow to

gaining streams downgradient from residential development may contribute OWCs to surface-water or nearby groundwater sources. These and other nonpoint, mixed-use areas have been implicated as potential sources of low-level OWCs in drinking water (Brown and Trombley, 2009). Other potential sources of contamination include manufacturing plants, hospitals, and confined agricultural feeding operations. Although WWTP effluents are only one potential source, human and animal wastewater effluent streams are among the most important sources of the majority of these compounds in the aquatic environment (Focazio and others, 2008).

Organic wastewater contaminants often originate from municipal wastewater discharge because many of them are not completely removed during wastewater treatment (Benotti and others, 2009). Runoff from chemically treated agricultural fields and concentrated domestic animal-feeding operations, groundwater inflow containing landfill leachates, and inflows from other nonpoint sources in urban areas may also contribute these contaminants to the environment (Barnes and others, 2004). Furthermore, some of these anthropogenic compounds persist through drinking-water treatment processes that were not designed to remove these contaminants. As analytical methods improve and detection limits and corresponding analytical reporting levels decrease into the nanogram-per-liter range, however, it is important to consider that the detection of pharmaceutical and other chemicals in drinking water may not imply a risk to human health (Benotti and others, 2009). More research is warranted to determine the potential risk associated with chronic exposure to trace concentrations of pharmaceutical and other unregulated contaminants in drinking water (Novak and others, 2011).

### Drinking-Water Withdrawals and Distribution in the Watershed

After Boston's water supply, the Merrimack River is the second largest surface-water source of drinking water in New England. Treated river water supplies 135,000 people through the LRWU; an additional 165,000 residents in Lawrence, Methuen, Andover, and Tewksbury, Mass., also use Merrimack River water. In New Hampshire, there is only one community drinking-water intake along the main stem. The Nashua drinking-water system pumps a portion of its water from the Merrimack River to supply the city and a few surrounding towns. Numerous farms and several municipal drinking-water well fields also pump water from the river. Most community water systems in New Hampshire rely on groundwater for their supplies (New Hampshire Department of Environmental Services, 2009). Demand for water resources will likely increase as the population increases in the watershed, and resource managers attempt to develop new sources to meet potable-water needs. Most of the aquifers in unconsolidated sand and gravel have already been developed for public supply, leaving surface water and bedrock aquifers as the only other viable sources for large developments in the future (Sarah Flanagan, U.S. Geological Survey, written commun., 2010).

## Sample Collection and Analysis

While the focus of this study was on the occurrence of pharmaceuticals in source water from the Merrimack River, the basic study design and methods of sample collection and analysis followed those of the larger USGS SWQA.

### Site Selection and Field-Data Collection

The Merrimack River was chosen as the site for the pharmaceutical investigation because it met specific criteria set out by the SWQA study design (Delzer and Hamilton, 2007). The SWQA site-selection criterion called for a surface-water source that is used to provide drinking water to a large population in the northeastern United States. The Merrimack River met this criterion in Massachusetts and New Hampshire. Additionally, the Merrimack River Basin extends over a large area and encompasses many land-use practices and potential sources of contamination; therefore, it was thought likely to carry some human, livestock and other agricultural, industrial, and wastewater contaminants to drinking-water intakes downstream. The SWQA program also investigates smaller drinking-water sources across the Nation—for example, the Hatfield, Mass., drinking-water source (Brown and Trombley, 2009).

Source- and finished-water samples were collected periodically from April 2008 through June 2009. Raw Merrimack River source water was sampled through an access door in the concrete floor of the LRWU intake facility enclosing a wet well at the edge of the river and processed onsite in a mobile USGS laboratory. Samples of the finished water were collected from a continuously running spigot in the laboratory of the LRWU in the main building of the treatment plant. Finished-water samples were collected about 5 hours after raw-water samples to match the expected time of travel through the treatment process and to collect both samples from the same parcel of water. Source and finished drinking-water samples were collected by using trace-level sampling protocols (Wilde and others, 2004).

### Sample Processing and Laboratory Analysis

Grab samples of raw source water were collected by lowering a tethered weighted-bottle sampler equipped with either a clean 1-liter (L) Teflon bottle or a new 1-L baked amber-glass bottle. About 10 L of raw water was composited in a 14-L Teflon churn splitter. The composite sample was mixed in the churn, and samples were split from the 10-L composite for each monthly sampling round. All water samples were

analyzed for physical characteristics (temperature, specific conductance, pH, and dissolved oxygen), *E. coli* bacteria, pharmaceutical compounds and metabolites. Results of analyses for only pharmaceuticals, bacteria, and field characteristics are reported here. Samples for determination of other SWQA analytes were collected, processed, and shipped together for analysis at the USGS National Water Quality Laboratory (NWQL) in Lakewood, Colo.; these data are reported in a separate USGS NAWQA publication (Carter and others, 2010).

Measurements of physical characteristics were made with a multiparameter water-quality monitoring instrument calibrated before use on the same day as sampling in accordance with manufacturer's instructions and USGS protocols (Wilde and others, 2004). For the raw-water measurements, the instrument was lowered to about 3 ft below the water surface inside the wet well; finished-water measurements were made by plumbing the flow from the laboratory tap through a cell attached to the instrument.

All raw-water samples were filtered through a 0.7-micrometer (µm) glass-fiber filter on a free-standing aluminum filter plate placed inside an isolation chamber. A Teflon diaphragm pump with C-Flex tubing was used to force the raw-water sample through the filter membrane into 1-L baked amber-glass sample bottles. Water pressure at the laboratory tap was sufficient to force finished-water samples through the plate filter. Finished water was collected in sample bottles containing 0.1 gram (g) of ascorbic acid and 7.8 g of rinsed Trizma pH-7 buffer. Free chlorine was reduced with the ascorbic acid, and samples were buffered to pH 7 to minimize acid-catalyzed hydrolysis of analytes during shipping and handling (Valder and others, 2008). Once the samples were collected in prelabeled, sterile, amber-glass bottles, they were placed in resealable bags, wrapped in foam packing material, inserted into large bags in plastic coolers filled with ice, chilled to 4 degrees Celsius (°C), and shipped overnight to the NWQL for analysis. Samples were analyzed for pharmaceuticals by solid-phase extraction onto chemically modified styrene-divinylbenzene resin, followed by high-performance liquid chromatography/mass spectrometry (Furlong and others, 2008).

Raw-water *E. coli* bacteria samples were collected in sterile bottles attached to the weighted-bottle sampler that was lowered through the floor into the wet well. Bacteria in the samples were enumerated by using modified mTEC agar and membrane filtration (U.S. Environmental Protection Agency, 2000). The process of sample plating was completed onsite inside the USGS mobile laboratory. The plates were then incubated for 24 hours, after which colonies on the plates were photographed and counted at the USGS Massachusetts-Rhode Island Water Science Center laboratory in Northborough, Mass.

# Quality of Finished Water

The QA plan for this study was designed with the primary objective of obtaining environmental data that are representative of the raw and finished water collected during the study period. Specific guidance for the collection of samples was obtained from the USGS National Field Manual for the Collection of Water-Quality Data (U.S. Geological Survey, variously dated).

## Quality-Assurance and Quality-Control Samples

The QA plan established protocols that were followed to ensure consistency throughout the project. In addition to consistent protocols, quality-control (QC) samples were collected to assess the data. Quality-control samples helped verify analytical results and consisted of four blank samples, two replicate samples, and two spike samples. Blank samples were collected to check for potential contamination in the sampling process and laboratory analysis. Replicate samples were used to check laboratory variability, and spike samples helped assess analytical accuracy.

Four blank samples were collected during the study: two field blanks and two source-solution blanks. Field blanks were collected in the same manner and setting as the environmental samples, except that deionized water was used in place of the environmental sample. Source-solution blanks contained nitrogen-purged organic-free deionized water. Blanks were always collected in bottles containing the dechlorination agent ascorbic acid to assess whether the ascorbic acid caused analytical interferences. No pharmaceutical analytes were detected in any of the blank samples (table 2), indicating that samples were not contaminated during the sampling process and laboratory analysis.

Replicate samples are additional samples that are designed to be identical in composition to the corresponding environmental samples. Replicate samples provide a measure of bias and variability for the methods of sample collection, processing (churning and filtering), and laboratory analysis, and for possible effects such as analyte degradation prior to laboratory analysis (Smith, 2008). Replicate samples were collected twice—once from raw, untreated source water and once from finished, treated drinking water. There were no measurable differences in concentrations of any analytes in any of the replicate samples (table 2). Caffeine and carbamazepine were detected in both the raw-water sample and its QC replicate, but the concentrations were too small to be reliably quantified.

To produce laboratory-spike samples, additional environmental replicate samples were collected, processed, and shipped to the NWQL. These samples were subsequently fortified at 0.25 micrograms per liter (µg/L) for all pharmaceuticals tested (table 2), and then analyzed. The percent recovery for each target analyte added to the environmental sample is used to determine bias and variability arising from

**Table 2.** Concentrations of pharmaceutical analytes in quality-control samples and ranges of percent recoveries of known concentrations of compounds added to samples.

[µg/L, micrograms per liter; <, less than; --, not measured]

| Quality-assurance sample type | Date | 1,7-Dimethyl-xanthine (µg/L) | Acetamino-phen (µg/L) | Albuterol (µg/L) | Caffeine (µg/L) | Carbamazepine (µg/L) | Carbamazepine-d10 (percent recovered) | Codeine (µg/L) | Cotinine (µg/L) |
|---|---|---|---|---|---|---|---|---|---|
| Raw water | Oct. 22, 2008 | <0.120 | <0.080 | <0.060 | <0.200* | <0.040* | 75.8 | <0.040 | <0.026 |
| Raw-water replicate | Oct. 22, 2008 | <.120 | <.080 | <.060 | <.200* | <.040* | 77.1 | <.040 | <.026 |
| Finished water | Oct. 22, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | 86.0 | <.040 | <.026 |
| Finished-water replicate | Oct. 22, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | 67.3 | <.040 | <.026 |
| Blank water | Apr. 16, 2008 | <.100 | <.080 | <.040 | <.060 | <.040 | 101 | <.040 | <.026 |
| Blank water | Nov. 19, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | 137 | <.040 | <.026 |
| Blank water | Nov. 19, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | 135 | <.040 | <.026 |
| Blank water | Jan. 8, 2009 | <.120 | <.080 | <.060 | <.200 | <.040 | 95.0 | <.040 | <.026 |

| Quality-assurance sample treatment | Date | 1,7-Dimethyl-xanthine | Acetamino-phen | Albuterol | Caffeine | Carbamazepine | Carbamazepine-d10 | Codeine | Cotinine |
|---|---|---|---|---|---|---|---|---|---|
| Finished water sample | May 21, 2008 | <.100 | <.080 | <.040 | <.060* | <.040 | 35.0 | <.040 | <.026 |
| Percent spike recovered with ascorbic acid treatment | May 21, 2008 | 68.5 | 47.7 | 40.7 | 112 | 124 | -- | 82.2 | 95.2 |
| Percent spike recovered without ascorbic acid treatment | May 21, 2008 | 69.4 | 49.3 | 12.4 | 129 | 101 | -- | 49.8 | 93.6 |

| Quality-assurance sample type | Date | Dehydronife-dipine (µg/L) | Diltiazem (µg/L) | Diphenhy-dramine (µg/L) | Ethyl nicotinate-d4 (percent recovered) | Sulfa-methoxazole (µg/L) | Thiabenda-zole (µg/L) | Trim-ethoprim (µg/L) | Warfarin (µg/L) |
|---|---|---|---|---|---|---|---|---|---|
| Raw water | Oct. 22, 2008 | <0.080 | <0.080 | <0.040 | 74.0 | <0.160 | <0.060 | <0.020 | <0.100 |
| Raw-water replicate | Oct. 22, 2008 | <.080 | <.080 | <.040 | 72.8 | <.160 | <.060 | <.020 | <.100 |
| Finished water | Oct. 22, 2008 | <.080 | <.080 | <.040 | 82.9 | <.160 | <.060 | <.020 | <.100 |
| Finished-water replicate | Oct. 22, 2008 | <.080 | <.080 | <.040 | 63.9 | <.160 | <.060 | <.020 | <.100 |
| Blank water | Apr. 16, 2008 | <.060 | <.040 | <.050 | 100 | <.100 | <.100 | <.040 | <.060 |
| Blank water | Nov. 19, 2008 | <.080 | <.080 | <.040 | 156 | <.160 | <.060 | <.020 | <.100 |
| Blank water | Nov. 19, 2008 | <.080 | <.080 | <.040 | 159 | <.160 | <.060 | <.020 | <.100 |
| Blank water | Jan. 8, 2009 | <.080 | <.080 | <.040 | 94.7 | <.160 | <.060 | <.020 | <.100 |

| Quality-assurance sample treatment | Date | Dehydronife-dipine | Diltiazem | Diphenhy-dramine | Ethyl nicotinate-d4 | Sulfa-methoxazole | Thiabenda-zole | Trim-ethoprim | Warfarin |
|---|---|---|---|---|---|---|---|---|---|
| Finished water sample | May 21, 2008 | <.060 | <.040 | <.050 | 32.5 | <.100 | <.100 | <.040 | <.060 |
| Percent spike recovered with ascorbic acid treatment | May 21, 2008 | 114 | 94.1 | 81.5 | -- | 132 | 59.3 | 84 | 97.4 |
| Percent spike recovered without ascorbic acid treatment | May 21, 2008 | 100 | 2.4 | 26.5 | -- | 36.6 | 59 | 45.8 | 22.7 |

*Analyte detected at a concentration that could not be reliably quantified.

(1) the degradation of target analytes during shipment to and holding by the laboratory, (2) the analytical method, and (3) interferences that mask or enhance determinations of the target analytes in the environmental sample or produce matrix effects (Smith, 2008).

In general, there was significant improvement in percent recovery of spiked compounds with the addition of ascorbic acid. Percent recoveries in the absence of ascorbic acid ranged from 2.4 for diltiazem to 129 for caffeine (table 2). With the addition of ascorbic acid, percent recovery of diltiazem increased to 94.1 and other low recoveries were also increased. Overall, percent recoveries in the presence of ascorbic acid, which was routinely applied to the finished-water samples, ranged from 40.7 for albuterol to 132 for sulfamethoxazole.

In addition to the QC samples collected during this project, the NWQL routinely analyzes other types of QC samples including laboratory-reagent blanks, interference-check solutions, laboratory control samples, standard-reference materials, laboratory-reagent spike samples, and laboratory duplicate samples to test and track method performance (Garbarino and others, 2006; Furlong and others, 2008). The NWQL also adds surrogate compounds (carbamazepine-d10 and ethyl nicotinate-d4) to all samples for routine determinations of percent recovery. Surrogate compounds are expected to react similarly to the targeted environmental analytes in the laboratory. Because these compounds are not normally found in the environment, the recovery of the surrogate compounds can be used to qualify the performance of the analysis (Smith, 2008). The percent recovery of carbamazepine-d10 in QA samples ranged from 35.0 to 137 (table 2), whereas the percent recovery of ethyl nicotinate-d4 in QA samples ranged from 32.5 to 159. The variations in recoveries observed in method surrogate compounds and in laboratory matrix-spike samples likely result from sample-specific matrix effects as well as losses during sample preparation and handling. The ranges of recoveries reported here are comparable to previously published observations for surface-water samples (Furlong and others, 2008).

## Pharmaceuticals and Water-Quality Constituents in Merrimack River Source Water

Samples were collected periodically from April 2008 through June 2009 and did not target any particular flow regime (fig. 2). Thus, Merrimack River water was collected and analyzed from flows that were above, below, and about average compared to long-term daily mean flow records. Physical characteristics were measured and bacteria samples collected during all 14 SWQA sampling rounds. Samples for pharmaceutical compounds were collected during 12 of the 14 rounds (fig. 2, table 3).

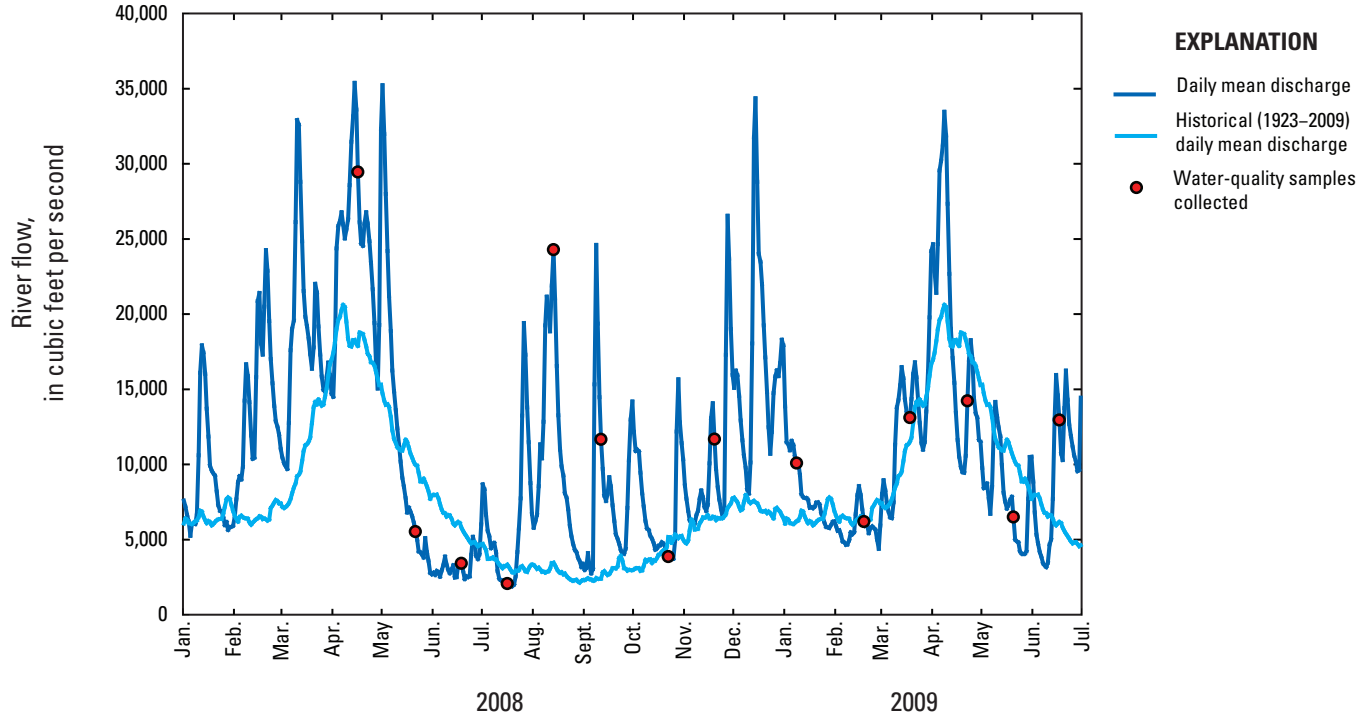


**Figure 2.** Daily mean river flow at the U.S. Geological Survey streamgage on the Merrimack River below the confluence with the Concord River at Lowell, Massachusetts, for January 2008 through July 2009, compared with historical mean daily river flows for the period June 1923 through September 2009, and times when samples were collected for chemical analysis.

**Table 3.** Analytical results for human-health pharmaceuticals measured in raw source water and finished drinking water at the Lowell Regional Water Utility, April 2008–May 2009.

[µg/L, micrograms per liter; <, less than]

| Sampling date | 1,7-Dimethylxanthine (µg/L) | Acetaminophen (µg/L) | Albuterol (µg/L) | Caffeine (µg/L) | Carbamazepine (µg/L) | Codeine (µg/L) | Cotinine (µg/L) | Dehydronifedipine (µg/L) | Diltiazem (µg/L) | Diphenhydramine (µg/L) | Sulfamethoxazole (µg/L) | Thiabendazole (µg/L) | Trimethoprim (µg/L) | Warfarin (µg/L) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Raw water | | | | | | | | | | | | | | |
| Apr. 16, 2008 | <.100 | <.080 | <.040 | <.060* | <.040 | <.040 | <.026 | <.060 | <.040 | <.050* | <.100 | <.100 | <.040 | <.060 |
| May 21, 2008 | <.100 | <.080 | <.040 | <.060* | <.040* | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| June 18, 2008 | <.100 | <.080 | <.040 | <.060* | <.040* | <.040 | <.026 | <.060 | <.040 | <.050* | <.100 | <.100 | <.040 | <.060 |
| July 16, 2008 | <.100 | <.080 | <.040 | <.060 | <.040* | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| Sept. 11, 2008 | <.100 | 0.084 | <.040 | 0.068 | <.040 | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| Oct. 22, 2008 | <.120 | <.080 | <.060 | <.200* | <.040* | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Nov. 19, 2008 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Jan. 8, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Feb. 18, 2009 | <.120 | <.080* | <.060 | <.200* | <.040 | <.040 | <.026* | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Mar. 18, 2009 | <.120 | <.080* | <.060 | <.200* | <.040 | <.040 | <.026* | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Apr. 22, 2009 | <.120 | <.080* | <.060 | <.200* | <.040* | <.040 | <.026* | <.080 | <.080 | <.050* | <.160 | <.060 | <.020 | <.100 |
| May 20, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Finished water | | | | | | | | | | | | | | |
| Apr. 16, 2008 | <.100 | <.080 | <.040 | <.060* | <.040 | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| May 21, 2008 | <.100 | <.080 | <.040 | <.060 | <.040 | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| June 18, 2008 | <.100 | <.080 | <.040 | <.060* | <.040* | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| July 16, 2008 | <.100 | <.080 | <.040 | <.060 | <.040* | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| Sept. 11, 2008 | <.100 | <.080 | <.040 | <.060 | <.040 | <.040 | <.026 | <.060 | <.040 | <.050 | <.100 | <.100 | <.040 | <.060 |
| Oct. 22, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Nov. 19, 2008 | <.120 | <.080 | <.060 | <.200 | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Jan. 8, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Feb. 18, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Mar. 18, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| Apr. 22, 2009 | <.120 | <.080 | <.060 | <.200* | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |
| May 20, 2009 | <.120 | <.080 | <.060 | <.200 | <.040 | <.040 | <.026 | <.080 | <.080 | <.040 | <.160 | <.060 | <.020 | <.100 |

*Analyte detected at a concentration that could not be reliably quantified.

Of the 14 prescription and nonprescription pharmaceutical compounds analyzed in raw Merrimack River source water, only 2 were detected at concentrations greater than their analytical reporting levels (RL). Concentrations of acetaminophen, a nonprescription analgesic (0.084 µg/L), and caffeine (0.068 µg/L) marginally exceeded the RLs in the September 2008 samples (table 3). There were additional detections of acetaminophen; caffeine; carbamazepine, an antiepileptic; cotinine, a metabolite of nicotine; and diphenhydramine, a nonprescription antihistamine, during the 14-month study (table 3), but none at concentrations large enough to be reliably quantified. The estimated concentrations were above the analytical-method detection limit but below the RL. The RL is defined as two times the long-term analytical method detection limit observed by the laboratory (Furlong and others, 2008). Adjustments to RLs are commonly made by the NWQL (for example, in October 2008 the RL for albuterol was changed from 0.040 to 0.060 µg/L) and are based on statistical quantification of the analytical-method performance. The goal is to maintain the rate of false-positive results at no more and no less than 1 percent. An explanation of how analytical RLs are determined is given elsewhere (Childress and others, 1999).

Physical characteristics and *E. coli* bacteria were also measured in raw-water samples, and the results compared to applicable standards. The data were consistently within the standards of the Commonwealth of Massachusetts and met criteria established by the U.S. Environmental Protection Agency (USEPA) for the physical characteristics measured (table 4). The USEPA issues guidance in the form of National surface-water-quality criteria for approximately 150 contaminants to protect aquatic life and human health. These criteria are published pursuant to Section 304(a) of the CWA and provide guidance for states and tribes to use in adopting water-quality standards (U.S. Environmental Protection Agency, 2009). The Massachusetts Department of Environmental Protection (MassDEP) issues surface-water-quality standards (Massachusetts Department of Environmental Protection, 2007) incorporating USEPA guidance for site-specific conditions. The intent of these standards is to support a designated-use classification, such as habitat for fish and aquatic life or a human use, such as primary-contact recreation (for example, swimming or fishing), secondary-contact recreation (for example, sport fishing or boating; fig. 3), or use as a drinking-water supply.

Raw-water measurements of pH were consistent with the MassDEP criterion for this segment of the Merrimack River: within the range of 6.5 through 8.3 standard units (table 4). The pH of finished water is adjusted in the treatment process to be slightly basic—in the range of 7.4 to 8.4 standard units— to aid in the coagulation, flocculation, and removal of suspended sediment and other impurities. An added benefit

**Table 4.** Physical characteristics and bacterial concentrations in samples of raw source water and finished drinking water collected monthly at the Lowell Regional Water Utility, April 16, 2008, through June 17, 2009.

[°C, degrees Celsius; µS/cm, microsiemens per centimeter; mg/L, milligrams per liter; CFU/100 mL, colony forming units per 100 milliliters; --, not sampled]

| Date | Temperature (°C) | | Specific conductance (µS/cm) | | pH (standard units) | | Dissolved oxygen (mg/L) | | *E. coli* (CFU/100 mL) | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Raw-water | Finished water | Raw water | Finished water | Raw water | Finished water | Raw water | Finished water | Raw water | Finished water |
| 2008 | | | | | | | | | | |
| Apr. 16, 2008 | 6.4 | -- | 76 | 155 | 7.1 | 8.0 | 10.4 | -- | 2 | -- |
| May 21, 2008 | 15.1 | 15.9 | 147 | 209 | 6.8 | 7.7 | 9.3 | 8.9 | 12 | -- |
| June 18, 2008 | 22.2 | 22.7 | 159 | 220 | 6.8 | 7.9 | 7.4 | 7.3 | 28 | -- |
| July 16, 2008 | 25.4 | 25.8 | 170 | 208 | 6.6 | 7.6 | 7.6 | 7.6 | 9 | -- |
| Aug. 13, 2008 | 19.1 | 20.2 | 75 | 156 | 6.7 | 7.8 | 7.0 | 8.3 | 190 | -- |
| Sept. 11, 2008 | 19.3 | 20.6 | 107 | 177 | 7.0 | 7.6 | 7.1 | 8.1 | 99 | -- |
| Oct. 22, 2008 | 15.1 | 14.1 | 143 | 206 | 6.9 | 7.7 | 10.8 | 11.0 | 18 | -- |
| Nov. 19, 2008 | 6.2 | 12.2 | 78 | 150 | 6.8 | 7.7 | 12.6 | 12.3 | 17 | -- |
| 2009 | | | | | | | | | | |
| Jan. 8, 2009 | 0.0 | 8.1 | 140 | 199 | 6.8 | 7.7 | 15.7 | 13.8 | 3 | -- |
| Feb. 4, 2009 | 0.4 | 4.4 | 160 | 220 | 6.7 | 8.0 | 15.4 | 15.1 | -- | -- |
| Feb. 18, 2009 | 0.5 | 4.8 | 141 | 192 | 6.6 | 7.8 | 12.5 | 13.4 | 2 | -- |
| Mar. 18, 2009 | 2.8 | 6.1 | 130 | 191 | 6.7 | 8.0 | 12.6 | 12.8 | 1 | -- |
| Apr. 22, 2009 | 10.0 | 12.2 | 128 | 206 | 6.7 | 7.8 | 10.6 | 10.8 | 25 | -- |
| May 20, 2009 | 16.0 | 17.6 | 123 | 194 | 6.5 | 7.8 | 8.8 | 9.1 | 13 | -- |
| June 17, 2009 | 17.9 | 19.0 | 98 | 164 | 6.5 | 7.7 | 8.2 | 8.3 | 120 | -- |

**Figure 3.** Kayakers paddling on the Merrimack River near Lowell, Massachusetts, June 2009.

of slightly basic drinking water in the distribution system is a reduction in the corrosion potential of residential plumbing, heating equipment, and water fixtures; heavy metals, such as lead in solder and copper in pipes, may dissolve under acidic conditions. In this study, the pH of finished-water samples was consistently within the range of 7.6 to 8.0 (table 4).

Dissolved-oxygen levels in the raw water always exceeded the MassDEP warm-water minimum of 5.0 milligrams per liter (mg/L) for this segment of the Merrimack River. The lowest dissolved-oxygen measurement (7.0 mg/L) was recorded in August 2008. It is interesting to note that this measurement also exceeds the cold-water minimum (6.0 mg/L) for this river. Temperature measurements were always lower than the maximum of 28.3°C for warm-water fisheries. The highest temperature of 25.4°C was recorded in July 2008, and the lowest was 0.0°C in January 2009 (table 4).

*E. coli* concentrations were determined for raw water collected at the intake. The largest *E. coli* concentration, 190 colony-forming units per 100 milliliters (CFU/100 mL), was measured in August 2008. The August 2008 result was below the Massachusetts criterion for a single sample collected at a site far from bathing beaches (235 CFU/100 mL maximum). Additionally, the criterion specifies that the geometric mean of all *E. coli* samples taken within the most recent 6 months shall not exceed 126 CFU/100 mL (based on a minimum of five samples). Following this calculation algorithm for the geometric mean, the *E. coli* concentrations measured in this study never exceeded 34 CFU/100 mL for six consecutive monthly samples. These relatively low *E. coli* counts are consistent with other water-quality results from this study, including the few detections of pharmaceutical compounds at reportable concentrations.

Annual variations in Merrimack River flow conditions were well represented in the study: sampling was conducted during high and low flows and in all seasons. The monthly sampling schedule, however, may have missed certain hydrologic events that could have transported greater concentrations of OWCs to the sampling site. Based on the size of the watershed, it is likely that runoff volume effectively dilutes any contaminant signal. It is also likely, however, that recent efforts to separate stormwater- and wastewater-discharge systems and the development of outreach programs to educate the public about the effects of nonpoint-source pollution and proper practices for the disposal of pharmaceuticals in the reaches upstream from the LRWU have greatly reduced the potential for contamination at the LRWU intake.

## Effects of Treatment on Pharmaceutical Compounds in Merrimack River Source Water

Municipal drinking-water treatment plants (DWTPs) are designed to remove contaminants and other objectionable materials from raw water through the treatment process. Most DWTPs use a combination of processes including coagulation, flocculation, sedimentation, primary disinfection, filtration, secondary disinfection, and fluoridation. The LRWU uses chlorination disinfection, flocculation, sand- and dual-media filtration, granular activated-carbon absorption, pH adjustment, and fluoridation. Chlorination and other oxidation-treatment technologies (such as ozonation) are effective at removing bacteria and other pathogens from drinking water. Additional benefits of chlorination (and ozonation) have been reported to include the effective removal (or transformation to oxidation byproducts) of some pharmaceuticals and other OWCs (Westerhoff and others, 2005). Westerhoff found that chlorine treatment oxidized more than 80 percent of the compounds tested in bench-scale experiments. These compounds were always oxidized more effectively by ozonation, but some compounds were poorly oxidized by either treatment. Westerhoff and others (2005) also reported improved removal of some OWCs by activated-carbon treatment.

Samples of finished water from the LRWU did not contain reportable levels of any of the pharmaceuticals measured in this study; however, two of the five compounds detected in corresponding raw-water samples (caffeine and carbamazapine) were detected in finished water at levels that could not be reliably quantified. Detection of these two compounds in the finished water samples may indicate that they are resistant to removal through the drinking-water treatment process, but could also be indicative of a false-positive result. Carbamazepine, in particular, has been observed to persist through drinking-water treatment processes (Stackelberg and others, 2004; Benotti and others, 2009). None of the other 12 pharmaceuticals was detected in any of the finished-water samples (tables 1 and 3). Recent advances in analytical methods have allowed quantification of pharmaceutical compounds at nanogram-per-liter

concentrations. The work of Benotti and others (2009) is notable in that analytical results are reported at concentrations below reporting levels of the methods employed by the USGS NWQL and reported on here. Our results are at tenths to hundredths of a microgram per liter reporting levels. It is possible that additional pharmaceutical compounds would have been detected using methods with nanogram-per-liter reporting levels.

No drinking-water standards or health advisories have been established for any of the pharmaceutical compounds detected in this study, and monitoring of these contaminants in the Nation's waters used for public supply is lacking. Furthermore, drinking-water criteria currently are based on the toxicity of individual compounds and not combinations of compounds. Little is known about potential human-health effects associated with chronic exposure to trace levels of multiple OWCs through routes such as drinking water (Stackelberg and others, 2004).

## Summary

The U.S. Geological Survey, in cooperation with the Massachusetts Department of Environmental Protection, examined the presence of 14 commonly used pharmaceutical compounds and fecal-indicator bacteria in Merrimack River water, which is used as a drinking-water source for more than 135,000 residents and businesses in the communities of Lowell, Dracut, Tyngsboro, and Chelmsford in eastern Massachusetts. Samples of the raw, untreated source river water were collected at the surface-water intake of the Lowell Regional Water Utility (LRWU), and the same parcel of water was sampled after treatment and before entering the distribution system (finished-water samples). Water samples were collected periodically from April 2008 through June 2009 and included 12 source-water samples paired with 12 finished-water samples, plus several quality-control samples. Additional source-water samples were collected for determination of concentrations of the fecal-indicator *E. coli* and measurements of several physical characteristics.

Despite many potential sources of contamination in the drinking-water source area, only 2 of the 14 target analytes were found at concentrations above the reporting level. Acetaminophen, a nonprescription analgesic, and caffeine were measured in the September 2008 source-water samples at concentrations of 0.084 and 0.068 micrograms per liter, respectively. Three other compounds—carbamazepine, an antiepileptic; cotinine, a metabolite of nicotine; and diphenhydramine, a nonprescription antihistamine—also were detected in source-water samples, but their concentrations were too low to be reliably quantified. Values measured for physical characteristics, including temperature, pH, specific conductance, dissolved oxygen, and *E. coli* bacteria, were always within regulatory guidelines.

None of the 14 pharmaceuticals was found in the finished water at a reportable concentration, defined as two times the long-term detection limit observed by the U.S. Geological Survey National Water Quality Laboratory. Two compounds, however, caffeine and carbamazepine, were detected in finished water, but at concentrations that were too low to be reliably quantified and could be considered as false positives. Physical and chemical characteristics of the finished water were comparable to those of the raw water and reflected adjustments made during the 5-hour treatment process. *E. coli* was not analyzed in finished water because disinfection during the treatment processes is effective at removing bacteria and other pathogens from the drinking-water supply; the LRWU also routinely conducts these analyses as part of their consumer-confidence protocols.

It is possible that the monthly sampling schedule failed to detect contamination resulting from storm events, although sampling did include both high and low flow conditions. It is also possible that contaminant signals were effectively diluted by the large discharge volumes at that point in the river. It is also likely, however, that reductions in pollution potential may have been realized through efforts to eliminate combined sewer overflows from several communities in the watershed, and that future plans to remediate existing combined sewer systems will continue to improve water quality at downstream reaches where the Merrimack River is used as the drinking-water source.

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For more information concerning this report, contact:

Director
U.S. Geological Survey
Massachusetts-Rhode Island Water Science Center
10 Bearfoot Road
Northborough, MA 01532
dc_ma@usgs.gov

or visit our Web site at:
http://ma.water.usgs.gov