

Article

Integrated Assessment of Shallow-Aquifer Vulnerability to Multiple Contaminants and Drinking-Water Exposure Pathways in Holliston, Massachusetts

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Abstract: Half of U.S. drinking water comes from aquifers, and very shallow ones (<20 feet to water table) are especially vulnerable to anthropogenic contamination. We present the case of Holliston, a Boston, Massachusetts suburb that draws its drinking water from very shallow aquifers, and where metals and solvents have been reported in groundwater. Community concerns focus on water discolored by naturally occurring manganese (Mn), despite reports stating regulatory aesthetic compliance. Epidemiologic studies suggest Mn is a potentially toxic element (PTE) for children exposed by the drinking-water pathway at levels near the regulatory aesthetic level. We designed an integrated, community-based project: five sites were profiled for contaminant releases; service areas for wells were modeled; and the capture zone for one vulnerable well was estimated. Manganese, mercury, and trichloroethylene are among 20 contaminants of interest. Findings show that past and/or current exposures to multiple contaminants in drinking water are plausible, satisfying the criteria for *complete exposure pathways*. This case questions the adequacy of aquifer protection and monitoring regulations, and highlights the need for integrated assessment of multiple contaminants, associated exposures and health risks. It posits that community-researcher partnerships are essential for understanding and solving complex problems.

Keywords: drinking water; shallow aquifers; exposure pathways; manganese (Mn); community based participatory research (CBPR)

1. Introduction

Safe drinking water is fundamental for public health, and while microbial contamination is a major concern globally, toxic chemical agents—both natural and anthropogenic—in water also pose health risks. Chemical contamination is particularly relevant for drinking water systems that draw from shallow surficial unconfined aquifers (<100 feet to water table) because they are vulnerable to

industrial contamination from waste releases to surface waters and/or soil, and are simultaneously frequently naturally enriched in metals (e.g., manganese (Mn), arsenic (As)) that release from confining bedrock. Half of U.S. drinking water comes from aquifers, and shallow ones are “very important sources for rural and small communities” [1].

In the U.S., metals such as Mn and mercury (Hg), as well as solvents such as trichloroethane (TCA) and trichloroethylene (TCE), are among common drinking water contaminants in aquifers [2,3]. Importantly, there is epidemiologic evidence that all four of these chemicals are neurotoxic individually, in particular following excess exposure that occurs prenatally or during early life [4–10]. Complicating this further is the potential for synergism between chemicals; however, little is known about how exposure to chemical mixtures impacts health [11]. The potential contribution of drinking water as a source of exposure to multiple chemicals is substantial: water is essential for life; many chemicals dissolve readily in water thereby increasing bioavailability; exposure can be acute or chronic in duration and can occur via multiple pathways (ingestion, inhalation and/or dermal/ocular contact); and entire populations may be at risk. Yet, research assessing exposures to and health effects of common drinking-water contaminants has been limited by inadequate sampling that fails to capture spatial/temporal variability, challenges in quantifying levels of multiple chemicals, and the lack of a biomarker that can retrospectively characterize exposures during relevant periods [12].

In this study, we examine the case of a typical U.S. suburban community where very shallow aquifers (<20 feet to water table, <50 feet to bedrock) supply all of the public drinking water, in a setting where common drinking-water contaminants such as solvents and metals have been reported in soil, soil water and groundwater since the late 1960s. The town of Holliston, Massachusetts represents an ideal opportunity to characterize exposures to natural and anthropogenic contaminants: nearly 100% of residents utilize drinking water supplied by very shallow aquifers; the area has been impacted by landfills, industry, and contamination events; the geology contains naturally elevated Mn that releases to groundwater; and community residents regularly raise concerns about discolored tap water. In this paper, we profile five sites to identify *chemicals of interest* that have been released. Service areas for each well in the network were modeled to estimate residential exposure to drinking-water contaminants that may have been captured by each well. We estimated the capture zone for one well of high concern due to its proximity to two waste sites with reported contamination events (including a mercury spill). Our preliminary characterization of complex exposure pathways is informing follow-up epidemiological research.

The paper is organized as follows: Section 2 provides an overview of regulations related to drinking water and waste-site cleanup in Massachusetts; Section 3 details the study design and methods used for ongoing data collection and modeling; Section 4 describes results to date; Section 5 presents recommendations for scientific research, policy and regulation, and technology; and Section 6 summarizes conclusions.

2. Background on Regulations for Drinking Water and Waste Site Cleanup

2.1. Safe Drinking Water Act (SDWA)

Under the Safe Drinking Water Act (SDWA), the U.S. EPA sets national health-based standards for drinking water quality to protect our nation’s drinking water sources from naturally occurring and anthropogenic contaminants [13]. These standards include maximum contaminant levels (MCLs) set for known risk agents (e.g., As, lead (Pb), TCE), yet there are uncertainties and emerging concerns about the safety of other common contaminants, such as naturally occurring Mn [12]. Mn is currently only loosely ‘regulated’ under SDWA for aesthetic reasons alone (secondary MCL 0.05 mg/L), and in 2004, the U.S. EPA issued a lifetime health advisory (HA; 0.3 mg/L), a 1-day and 10-day HA for acute exposure (1.0 mg/L), and a special HA for infants under 6 months old (0.3 mg/L) [14]. However, the HA “does not mandate a standard for action; rather it provides practical guidelines for addressing Mn contamination problems” [14]. Globally, the World Health Organization (WHO) has recognized Mn as

a potential risk agent since 1958, when WHO International Standards for Drinking Water suggested that levels >0.5 mg/L would “markedly impair the potability of the water” [15,16].

The SDWA allows for states to implement the provisions of the SDWA within their jurisdiction, provided that they can demonstrate that they have or will adopt standards as stringent, or more stringent, as those set by the U.S. EPA [13]. In Massachusetts, the Massachusetts Department of Environmental Protection (MDEP) regulates drinking water to meet the requirements of the SDWA. MDEP has the authority under Massachusetts General Law, Chapter 111, Section 160, and under 310 CMR 22.00 of the Massachusetts Drinking Water Regulations to require compliance with “guidelines” (the use of the term “guidelines” in the context of regulatory compliance sends a message that there is flexibility in the ways to be considered “compliant”). The findings of the Source Water Assessment and Protection (SWAP) program (see Section 2.2) can be used to recommend actions to further protect aquifers, but again it connotes an undesirable flexibility in regulation. The relatively high autonomy of towns in New England on policy matters adds another degree of problematic freedom to regulation of public water systems (PWSs), including design, installation, source approval, treatment and chemical application, water storage and distribution, and operation and monitoring procedures [17].

2.2. Source Water Assessment and Protection (SWAP) Program

The 1974 U.S. SDWA originally focused primarily on treatment as the means to provide safe drinking water to consumers at the point of consumption. In 1996, amendments to the SDWA established the Source Water Assessment and Protection (SWAP) Program to emphasize source water protection as a pollution prevention measure [18]. Under SWAP, each state is required to develop their own SWAP program to examine existing and potential threats of contamination to its public drinking water supply resources. For Massachusetts aquifers, the SWAP program characterizes the “susceptibility” of an aquifer to contamination by gathering information about industrial, commercial and land-use activities within aquifer recharge areas and capture zones for existing or proposed wells [17]. SWAP findings can be used to “recommend” local actions to further protect drinking water resources [17]. SWAP assessments had been carried out for all Massachusetts public water supplies by 2004 using the following steps:

1. Delineation of protection areas for all public groundwater and surface water sources;
2. Land use inventory to identify existing and potential threats to the quality of drinking water resources;
3. Determination of the susceptibility of the water supply resources to contamination from existing and potential threats; and
4. Publication and distribution of results to the general public.

Key to understanding, then responding to, the vulnerability of wells to contamination is setting buffer protection zones: “Zone I: the area closest to a well; a 100–400 foot radius proportional to the well’s pumping rate. This area should be owned or controlled by the water supplier and limited to water supply activities. Zone II: the primary recharge area for the aquifer. This area is defined by hydro-geologic studies that must be approved by [M]DEP. Zone III (the secondary recharge area) is the land beyond Zone II from which surface and ground water drain to Zone II and is often coincident with a watershed boundary” [18]. Zone II, representing the land surface area that contributes water to the well under severe pumping and aquifer recharge conditions, is non-trivial to establish because it requires modeling the well’s capture zone and recharge area under these conditions [19]. The capture zone is the three-dimensional aquifer volume whose groundwater is captured by the well as it pumps; the recharge area is the surface area of land for primary recharge by precipitation then infiltration. Capture-zone contamination represents direct contamination of the well’s drinking water supply either by contaminant release into it, or transport of contaminants by runoff and/or groundwater flow to it. Recharge-area contamination, either by contaminant release into it by infiltration/recharge or

contaminant transport by runoff to it, contaminates the larger aquifer body. This is further complicated by the fact that recharge areas for shallow unconfined aquifers can be quite large—the entire permeable area above the aquifer—whereas capture zones tend to be smaller and localized at the well as a function of pumping rate and aquifer characteristics. In confined aquifers that tend to be deeper, the recharge area can be far-removed from the area around a well, e.g., an upland area where aquifer layers outcrop at the surface can recharge an aquifer being exploited at depth in a lowland area. The SWAP report for the Town of Holliston public water supply (PWS #2136000) was completed in 2002 [18]; key findings are summarized in Section 3.1.

2.3. Waste Site Cleanup Regulations

In Massachusetts, cleanup of hazardous waste sites and spills are performed in accordance with the Massachusetts Contingency Plan (MCP), an extension of Massachusetts General Law Chapter 21E, and are administered by the MDEP [20]. MCP was promulgated in July 1993, and became effective October 1993. MCP established a regulatory framework under which those responsible for pollution are required to assess and remediate, as necessary, spills of oil and/or hazardous material (OHM) until cleanup criteria and risk assessment standards have been met [20]. If a spill or release of OHM exceeds the applicable reporting quantities listed in the MCP, the responsible party must first report the release to the MDEP then conduct an initial site investigation to determine if further investigation or remediation is required. Throughout the cleanup process the polluter is responsible for evaluating environmental conditions to determine if exposure pathways are complete or incomplete, with respect to human health risk (see Section 6).

3. Design and Methodology

3.1. Study Area and Drinking Water Supply

Holliston, Massachusetts (population 14,525, [21]) is located 21 miles west of Boston and is considered an ascendant bedroom community of the Boston Metrowest Region. It has an area of 19 sq. miles with ~5000 homes that are a mix of newer high-end development homes and older ranch-style homes. In 2010, 95% of the population was white, 27% was younger than 18 years old, and only 4% was in poverty [22].

Holliston operates its own public water supply and utilizes two local glacial overburden aquifers as its drinking water sources: the Bogastow Brook Aquifer (BBA) supplies water to the eastern portion of Holliston, while the Cedar Swamp Aquifer (CSA) supplies the western portion [19]. Both Zone IIs include wetland recharge areas; Holliston used to be the northern-most edge of the cranberry bog industry. CSA is recharged by Cedar Swamp wetland that overlies it, and its water table is mostly at the surface. Based on available information, we estimate that wells #1–5 were installed in the late 1960s/early 1970s, well #6 was installed in the 1990s as the population increased, and wells #7 and 8 were installed in 2014 and 2016, respectively. In 2016, five wells (#4, 5, 6, 7, 8) were used to withdraw groundwater for public drinking water (Figure 1). Well #8 was added at the same location as #2, and seems to have replaced it. All wells are gravel-packed, only 25–40 feet to base of extraction pipe. There are two water treatment plants, located at wells #4 (CSA) and #6 (BBA). Water treatment involves Mn and iron (Fe) filtration (Supplemental Material), disinfection with sodium hypochlorite, and corrosion control with zinc orthophosphate [23]. A network of pipes connects wells to homes and businesses, and are comprised of differing materials (asbestos cement, ductile iron, steel, polyvinyl chloride), diameters (6-, 8-, 10- and 12-inch), and ages. There are also three storage tanks and five to eight monitoring sites. Holliston lacks centralized wastewater treatment; each home or business has its own septic tank.

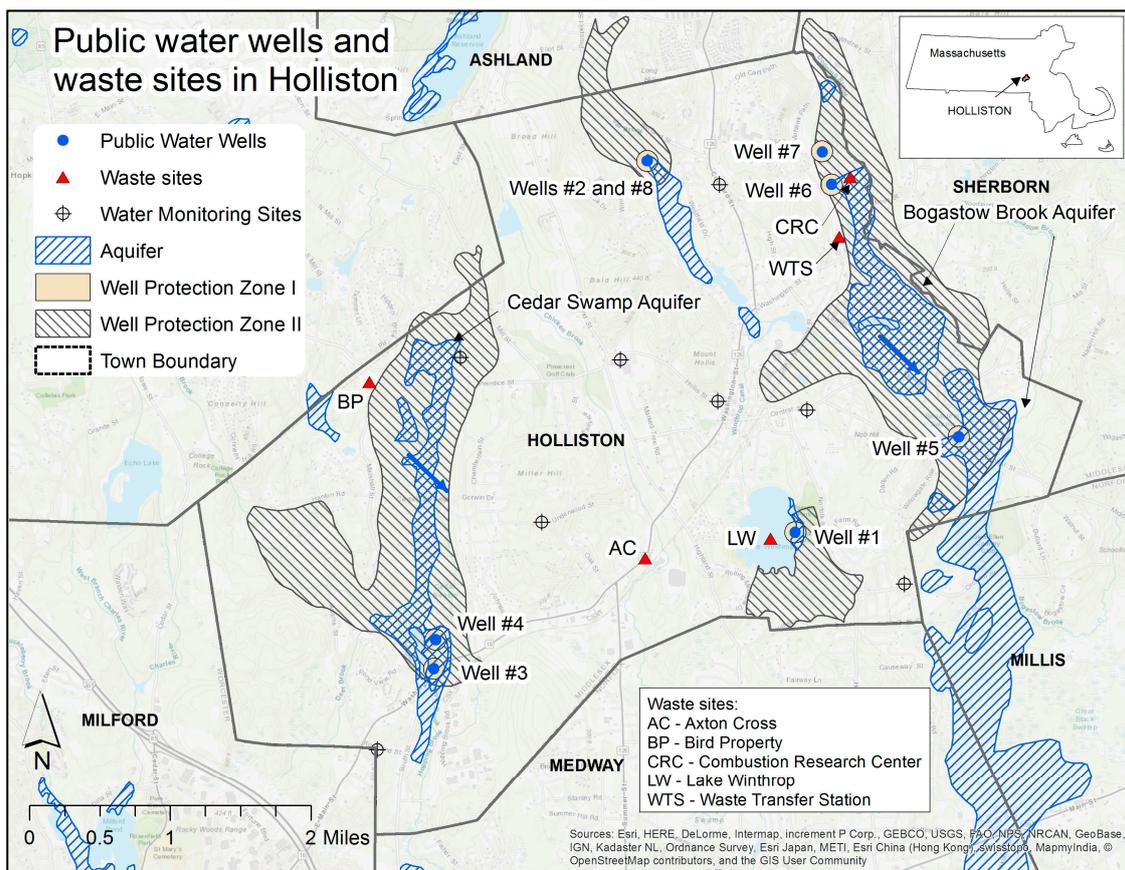


Figure 1. Public water wells and waste sites in Holliston, MA. Blue arrows indicate predominant ground water direction. Data sources: Bureau of Geographic Information (MassGIS), Commonwealth of Massachusetts, Executive Office of Technology and Security Services, Holliston Water Department.

In New England towns such as Holliston, local policy makers have considerable power and autonomy to make important decisions regarding public health and water. In Holliston, the local government is comprised of an elected Board of Selectmen (three members) chaired by the Town Administrator. The town has its own Board of Health, and the Department of Public Works manages the town drinking water supply. There are regular Town Meetings attended by residents who can raise concerns and comment on proposed policies and plans, including investments in water infrastructure. There have been numerous reports from residents of discolored, sometimes odorous tap water, an indicator of moderate to high Mn and/or Fe. Residents also express their concerns about drinking water quality via social media, exchanging posts and photographs of their tap water (Figure 2). One frequently asked question on the town website is: “Can I drink the water when it is colored?” The official response is: “The water has been treated, tested and is safe for consumption” [24]. Annual water quality Consumer Confidence Reports (CCRs) have stated that “iron and manganese are often present in groundwater at levels that can discolor the water or cause it to have unpleasant odors and tastes. Even though the water may be safe to drink, it is preferable that the iron and manganese be treated” [23].



Figure 2. Photos of discolored tap water in Holliston homes, posted on Facebook[®] by Holliston residents.

3.2. Integrated, Community-Based Approach to Drinking Water Research

Coupled human–environment systems are inherently complex in terms of constituent components, their makeup and behavior, dynamic interactions among them, and their temporal and spatial variability. The drinking water exposure scenario in Holliston is an example of this, and thus requires an integrated approach to research: thorough evaluation of potential health risks from (aquifer-based) drinking water involves addressing a series of questions using expertise from multiple disciplines and data sources (Table 1). For example, because chemicals move dynamically through the environment, including through aquifers, adequately understanding how drinking water quality varies in time and space is necessary to estimate health risks, which necessitates input from hydrogeologists, environmental chemists, engineers, exposure scientists, and health scientists. Further, comprehensive drinking water exposure assessment requires data integration as each data source by itself is inherently limited: secondary water monitoring data typically provide only sparse historic exposure information;

supplemental tap water sampling can fill in data gaps, but does not incorporate individual variability in absorption; and biomarker data reflect internal dose but not intake concentrations, and typically represent only narrow exposure periods. The use of a GIS platform facilitates data integration and permits spatial analyses of diverse types; health outcomes, non-genetic, place-based vulnerability/ risk factors, and exposure data are all spatially explicit [25], so GIS can detect important spatial patterns and associations otherwise obscured.

Table 1. Integrated approach to aquifer-based health risk research.

| Questions of Interest | Disciplines/Methods | Data Type/Analysis | Anticipated Product |
|---|--|---|---|
| What are local concerns and needs? What is the local knowledge? | Community-based participatory research (CBPR) | QL, QU, ET, CS—participatory | Compilation of reports |
| What are the characteristics of the aquifer? | Hydro-geochemistry; hydrology; GIS | QT, QL, GS, FL | Aquifer characterization |
| How is the aquifer exploited by water system, and how large are capture zones and service areas for each well? | Water engineering (hydraulics); GIS | QT, GS | Water supply system description |
| What are the sources and agents of aquifer contamination? How vulnerable is aquifer to contamination? | Environmental chemistry (fate and transport); risk analysis; GIS | QT, QL, GS, FL | Site profiles, List of contaminants of concern, Aquifer vulnerability rating |
| Who is exposed and what are the pathways of exposure? Are pathways complete? Among the exposed, who is most vulnerable? | Exposure science; risk analysis; GIS | QT, QL, GS, FL, QU, CT, ET | Exposure and risk profiling |
| What is the magnitude and timing of historic and current exposure(s)? | Exposure science; environmental epidemiology | QT, QL, GS, FL, QU, ET | Exposure findings and model |
| What are the associations between exposures and health outcomes? | Environmental epidemiology; GIS | QT, QL, QU, CT, ET | Epidemiologic findings |
| How are parameters of interest to participants and other stakeholders distributed spatially? | Participatory GIS | GS, e.g., photos of tap water by residents are mapped | Web-based information collection interface (“Holliston Health Atlas”), populated with data from participants (QA/QC by researchers) |
| If risks are elevated, how can they be reduced or mitigated? | All disciplines listed above (for source control, exposure reduction, adaptation/intervention) | PS informed by data from above | Risk communication and management plan. Water supply and treatment plan |

Key: QT—quantitative; QL—qualitative; GS—geospatial/GIS; FL—field sampling and lab tests; QU—questionnaire; CT—clinical testing; ET—ethnographic/narrative; CS—citizen science; PS—policy analysis and scenario modeling (alternative water sources, treatment options).

To maximize the likelihood that such integrated research meets community needs and is translated into actions to reduce vulnerabilities and health risks, a *vibrant partnership* between residents, researchers, local government and regulators is essential. Our study in Holliston was community initiated: in 2014, three parents (A.D., M.R., N.C.) contacted Clark University researchers (T.J.D., Y.O-H.) to share their concerns about known contamination sites in the town and problems with discolored tap water. They also pointedly posed questions about possible connections between drinking water quality and the health issues of their own children and their neighbors’ children. From this initial meeting grew a partnership between a group of eight to ten residents of Holliston and researchers, where mutual respect was fostered, co-generation of knowledge was encouraged, and project ownership was shared. Approximately four meetings per year have been held since 2014 in the home of a community member, where informal discussions between residents and researchers center on community priorities, questions, and strategies for addressing concerns. Stakeholders enjoy differentiated roles and responsibilities, but effort and reward are equitable. Community-based participatory research (CBPR) recognizes the ethical and functional importance of building strong communication and interactions

between researchers and other stakeholders, especially frontline communities most impacted by the problems under investigation [26–31]. An increasing number of federally supported environmental health research programs call for vibrant “bidirectional” community-researcher interactions from project inception through design, implementation and translation of findings to support interventions to improve health.

3.3. Waste Site Profiles and Identification of Contaminants of Interest

Through community dialogue, five sites of concern were identified that represent concerns expressed by residents about contamination: Waste Transfer Station (WTS); Combustion Research Center (CRC); Axton Cross (AC); Bird Property (BP); and Lake Winthrop (LW) (Figure 1). We mined secondary data related to these sites, including MDEP reports and contractor site assessments over the operating lifetimes of the sites (20–50 years) and/or during ongoing site clean-up in response to spills, accidental releases, MCP reporting needs and public concerns. Site profiles focused on the release of toxic chemicals and measurements of contaminant levels in environmental media. Considering potential health risks, we compiled a global list of *contaminants of interest* that comprised chemicals released or detected in environmental media.

3.4. Aquifer Characterization and Assignment of Vulnerability Ratings

We characterized the aquifer system in terms of its properties, boundaries, and Zone I, II and III buffers using secondary data from site assessment reports and field studies [18,19]. We then used published literature on vulnerability assessments of aquifers [32–34] and populations at risk from multiple health stressors [25] to construct a simple first-order vulnerability rating scheme, readily applicable to other populations/aquifers. It includes dimensions of contamination, exposure to risk agents/hazards, as well as the capacity to respond and adapt to reduce risks [35]. We rated each of 12 criteria on a 0–3 scale corresponding to zero; low (1); moderate (2); and high (3) vulnerability, based on secondary data from waste site field assessments (see Section 4.1), the 2002 SWAP Report [18], and dialogue with residents. Criteria were equally weighted, and ratings summed and divided by the total possible points for a worst-case scenario ($12 \times 3 = 36$), yielding an overall rating of 0.0–1.0 (1.0 = highest vulnerability).

3.5. Modeling Well #6's Capture Zone

The vulnerability of a well should consider the full capture zone of the well [34]. Capture-zone contamination leads directly to drinking water contamination and potential health risks. Pumping by a well alters the flow field in the vicinity of the well by bending streamlines; those within the capture zone are included, while those adjacent to it are bent towards the well (which acts as a point sink). Pumping can reverse the natural direction of groundwater flow just down gradient of the well, and the size of the capture zone is proportional to the pumping rate. Estimates of capture zones are therefore fundamental to the protection of drinking water source waters and delineating Zone II buffer protection zones [18].

We modeled the capture zone for Well #6 due to its proximity to contamination events and to two waste sites/area sources (WTS, CRC) (Figure 3). We used a two-step approach: (1) preliminary analytical modeling was conducted using established hydrogeological methods [36–38]; and (2) visual analytic element modeling (Visual AEM) was conducted, which involves single and multi-layer analytic element modeling of steady state groundwater flow and modeling of vertically averaged contaminant transport [39]. Input parameters included pumping rate, obtained from the Holliston Water Department; and aquifer characteristics (aquifer thickness, hydraulic conductivity, hydraulic gradient), obtained from site assessments (see Section 4.1). In the visual AEM, we accounted for uncertainty in hydraulic conductivity and variability in pumping rate.

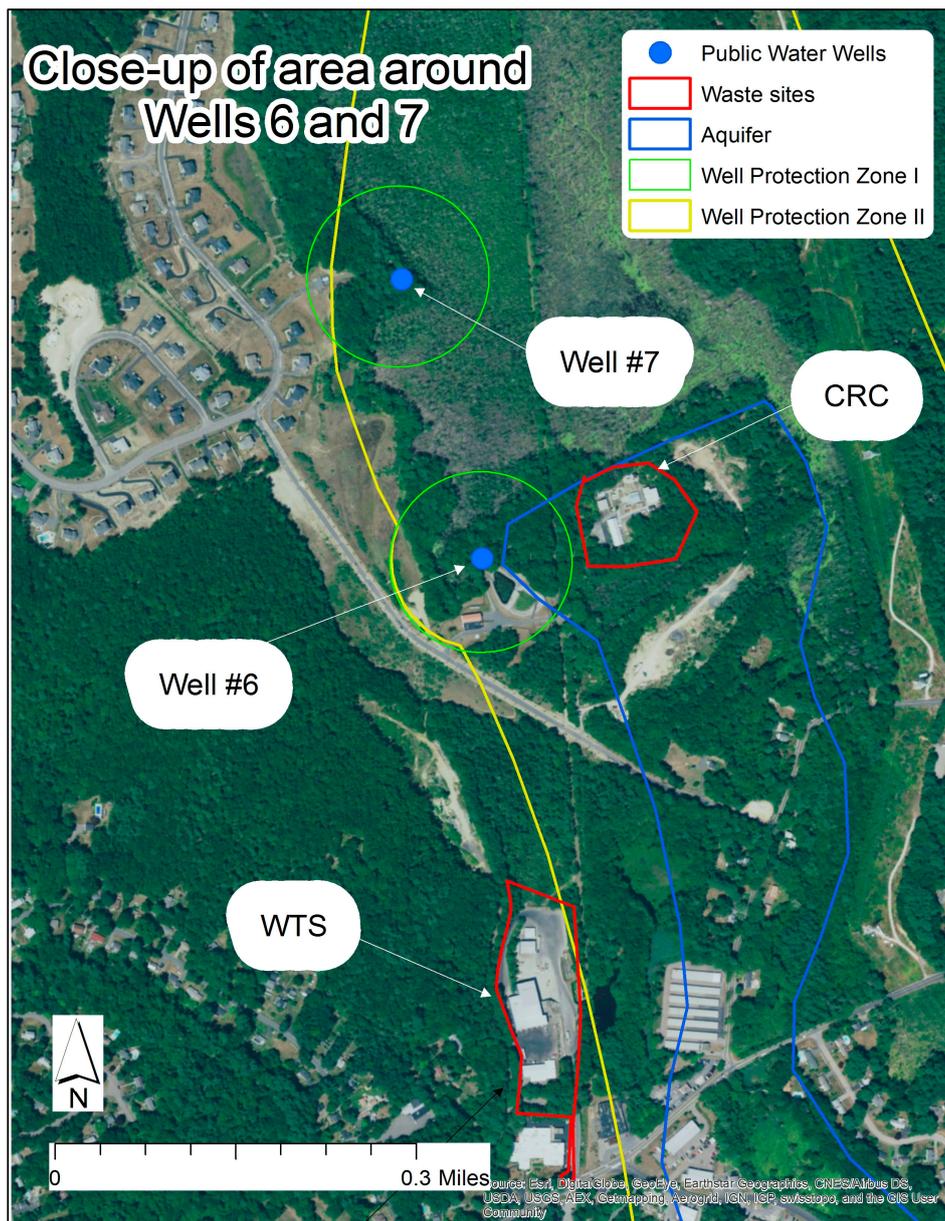


Figure 3. Close-up of area around Wells #6 and #7. Data sources: Google Earth® (from September 2014), Bureau of Geographic Information (MassGIS), Commonwealth of Massachusetts, Executive Office of Technology and Security Services. The housing development at top left is a newer (Phase I in 2000, Phase II completed 2016) high-end residential neighborhood, Holliston Heights. Location of wells outside the aquifer boundary suggests boundaries may need to be revised.

3.6. Modeling Well Service Areas

We obtained a map of the drinking water system via a Freedom of Information Act (FOIA) request made of the Town of Holliston in 2015. We created a digital model of the well/pipe network including pipe lengths and materials (for frictional losses), and estimates for each well's pump elevation. Elevation head was estimated using a digital elevation model (DEM) in ArcGIS®. Friction, elevation and pressure heads were summed to determine total dynamic head (TDH) required of each well/pump. The service area of each well was determined using reverse-engineering: initially a service area was assumed at a radius of 500 m for each well and the corresponding head losses and frictional losses were estimated using Hazen-Williams equation, with head losses due to elevation calculated by considering

the change in elevation of the pipes [40]. We were not able to obtain the make/model of installed pumps, so we chose a suitable candidate pump—Gould® (Xylem, Inc., Rye Brook, NY, USA) pump model #7TLC 550 GPM—as a surrogate. As a starting point, we calculated required TDHs at 500 m radii from each well then compared them with the pump’s rating curve (TDH vs. flow rate) for average and maximum flow rates for each well from 2000 to 2014. If the calculated TDH for a given well’s 500 m-radius service area was less than the pump’s TDH capability, then the radius of the service area was increased iteratively until required TDH was equal to or greater than TDH of the pump at average and maximum flow rates.

4. Results

4.1. Site Profiles

Findings are summarized in Table 2.

Table 2. Sites of concern, their contaminants of interest, and status.

| Site Name | Active or Legacy | Period (Years) | Operations | Contaminants of Interest Released at Site and/or Found in Site Assessments of Soil, Soil Water, Wetland Sediment and/or Groundwater | Site Status |
|----------------------------------|------------------|---------------------|--|--|------------------------------------|
| Waste Transfer Station (WTS) | Active | 1971–now (45) | Receipt, temporary storage and transfer of solid wastes | Total petroleum hydrocarbons (TPH); heavy metals such as lead, arsenic, and chromium; volatile organic compounds such as acetone and BTEX; semi-volatile organic compounds (SVOCs) such as benzo(a)pyrene, benzo(b)fluoranthene, chrysene, and 2-methylnaphthalene; and PCBs. | On-site monitoring |
| Combustion Research Center (CRC) | Active | 1968–now (50) | Outdoor testing of flame retardants, fire extinguishing agents | Mercury, manganese, cadmium, lead, PCE, TCE, DCE, trihalomethanes (chloroform, bromodichloromethane, dibromochloromethane), PBDEs, beryllium, barium and antimony trioxide. | On-site monitoring |
| Axton Cross (AC) | Legacy | 1967–1994 (27) | Chemical distribution and mixing | Solvents: methylene chloride, 1,1,1 trichloroethane, trichloroethylene, tetrachloroethene, 1,1 dichloroethane, 1,1 dichloroethene, trans 1,2 dichloroethene; vinyl chloride; p- and o-xylene; chloroethane, 4-isopropyltoluene, and 1,2,4-trimethylbenzene. Spill events: no. 2 heating oil (1976), xylene (before 1980), methylene chloride (1985), hydrofluoric acid (1984). | Superfund |
| Bird Property (BP) | Legacy | 1960s–1980s (20–25) | Illegal landfill for wastes. | Acetone, benzene, tetrachloroethene (PCE), trichloroethylene (TCE), 1,2-dichloroethane (DCA), toluene, cadmium. | Unclear: “no sig. risk” not issued |
| Lake Winthrop (LW) | Sources unclear | 1980s or earlier | Ecology, recreation. | 2,3,7,8-TCDD and mercury detected in fish and sediment samples (1984, 1996/7, 2012). | Sporadic testing. |

4.1.1. Waste Transfer Station

The Waste Transfer Station (WTS; 123 Washington Street, Holliston) is a municipal solid waste transfer station, collecting from institutional clients in the area. WTS was originally established for processing recycled materials in 1971, reassigned to dispose of solid waste in 1972, then transitioned to service as a waste transfer station in 1977. WTS is situated on the edge of Zone II for Well #6 (Figures 1 and 3).

In 1986, a notice of noncompliance was served, triggered by unfiltered drainage discharge. Since 1986, a total of 23 contaminants were detected at levels above MCP benchmark values in soil, wetland sediments, surface water, or groundwater. Most contaminants were detected during the site assessment response to a diesel fuel spill from an underground storage tank in 1988. Between 1988 and 1999, various site assessments reported measurable levels of contaminants, including total petroleum hydrocarbons (TPH); metals such as Pb, As, and chromium; volatile organic compounds such as acetone and BTEX; semi-volatile organic compounds (SVOCs) such as benzo(a)pyrene, benzo(b)fluoranthene, chrysene, and 2-methylnaphthalene; and polychlorinated biphenyls (PCBs) [41–52].

Data from a seismic refraction survey [49] and from the SWAP report [18] confirm aquifer vulnerability to contaminated runoff and spills from WTS. Facilities were upgraded in 2003–2004 to monitor wastewater discharge every month, and to control the odor in the adjacent wetland. In March 2004, in response to concerns from Holliston’s Conservation Commission, the Holliston Board of Health conducted an analysis of a “dense, black liquid” with an unusual odor being emitted from WTS drainage pipes. Unidentified petroleum hydrocarbons and lead were measured at concentrations of 9.31 mg/L and 0.11 mg/L, respectively. Notably, measured Pb levels were approximately ten times the MCL action level of 0.015 mg/L. Based on groundwater sampling data from WTS site assessments in 1999–2000, benzo(a)pyrene was detected at 0.5 µg/L (MCP GW-1 benchmark = 0.2 µg/L; SDWA MCL = 0.2 µg/L). However, no remedial actions were mandated given that this was considered wastewater that was not flowing directly into a surface-water drinking water source [53]. However, WTS acts as an area source of contaminated runoff that impacts a drinking water well’s Zone II (Figure 3), suggesting SWAP should be strengthened to protect aquifers from such area sources.

4.1.2. Combustion Research Center

The Combustion Research Center (CRC; 90 Brook Street, Holliston), owned by Kidde–Fenwal Inc., tests fire-extinguishing and flame-retarding equipment/materials for institutional customers. The mixed indoor and outdoor testing facility borders the U.S. Army Corps of Engineers-owned wetland area. The wetland is at a lower elevation than the site, with highly permeable topsoil and moderately permeable soil from about 5–25 feet deep [54]. Well #7 is situated in this wetland, and Well #6 is located only 400 feet from the CRC (i.e., at the edge of its Zone I) (Figures 1 and 3). Since 1968, the site has been a test center for flame retardants, including poly-brominated diphenyl ethers (PBDEs, suspected endocrine disrupting compounds) and fire extinguisher solutions (sodium or potassium bicarbonate, ammonium phosphate, antimony trioxide, argon/nitrogen “Argonite”, or CO₂) [55].

Key contaminants detected in soil, wetland sediments and groundwater on site and directly to the south include: Hg, Mn, cadmium, Pb, tetrachloroethylene (PCE), TCE, dichloroethene (DCE), trihalomethanes (chloroform, bromodichloromethane, dibromochloromethane), PBDEs, beryllium, barium and antimony trioxide [41,54,56]. Notably, an elemental Hg spill was reported in 2005 that impacted the adjacent wetland area and Dopping Brook. Site inspection by a private consultant revealed elemental Hg concentrations of 9.1 mg/L in surface water, which is more than 4500 times the MCL set by the U.S. EPA (MCL = 0.002 mg/L) [41]. United Technologies Corporation, from whom the property was leased at the time of the spill, remediated the wetland within 12 months through excavation and replacement with new sediment. Levels of TCE, DCE and PCE exceeding MCLs have also been reported in CRC monitoring reports [41,54,56]. Although PBDEs were not frequently detected at concentrations exceeding their respective MCLs, chronic contamination is a concern because they are persistent, toxic, and can bioaccumulate ($\log_{10} K_{ow}$ 6.2–6.6) [57,58].

Multiple notices of noncompliance were served by MDEP in the 2000s for inadequate testing and reporting. Following the Hg release in 2005, independent surface water testing was carried out and indicated insufficient site evaluation by CRC itself [59]. Available information suggests that Kidde-Fenwal currently reports quarterly data to MDEP from two on-site monitoring wells.

4.1.3. Axton Cross

Axton Cross (AC; 1 Cross Street, Holliston) is the site of a former distributor for textile, paper, rubber, metals, and plastics manufacturers. Operations at this site occurred between 1967 and 1994. It is listed with the MDEP as a disposal site under release tracking number (RTN) 2-00059 and is registered as a U.S. EPA Superfund Site #MAD059706937, though it is not currently on the National Priorities List (NPL) [60]. Adjacent surface water includes Chicken Brook and a wetland that feeds into it. Original construction altered the placement of the brook, and the now-buried original flow is

a potential source of groundwater discharge. Nearby surface water is 4–7 feet lower than the property, making it vulnerable to any runoff not intercepted by a wastewater treatment system.

In the 1970s, heating oil leaked into groundwater for an unknown period of time. A water–oil separation system was installed in 1976, and treated water was released into Chicken Brook. In the mid-1980s, 278 gallons of hydrofluoric acid and 670 gallons of methylene chloride were released into the environment, both due to equipment failure. Axton Cross was determined in the 1990s to be “the major contributor” to groundwater contamination documented by the MDEP. Concentrations of TCE in Chicken Brook were higher downstream of the facility than upstream, suggesting contribution of TCE from site activities [61]. In 2002, non-aqueous phase liquids (NAPLs) were detected in groundwater. The former leach field and discharge pit, wastewater treatment lagoon, two tank farms, underground storage tanks, and drum storage area on site were all identified as areas of concern. Contaminants of interest include DCE, PCE, and TCE in surface water and groundwater [61–69].

Axton Cross conducted extensive, costly investigations, including soil, surface water, and ground water sampling and analysis as well as remediation, although cleanup efforts were consistently delayed due to lack of funds. In 2006, AC Co. was approved for financial inability status by the MDEP, which allowed the company to defer remediation actions, but did not exempt it from potential liability. Thirteen monitoring wells have been installed on the site, but it is unknown when adequate remediation will be reached.

4.1.4. Bird Property

The Bird Property (BP; 708 Prentice Street, Holliston) is a former illegal landfill that operated during the 1960s–1980s between the towns of Holliston and Hopkinton. One small surface water pond and four patches of wetland upstream from Cedar Swamp occupy pieces of the property. Two public supply wells (#3 and 4) are located within a two-mile radius of BP (Figure 1). As delineated by the Holliston SWAP report, the majority of this site is CSA’s Zone III area [18], at the edge of Zone II. Runoff from BP can impact Zone II, and localized groundwater contamination at BP is hydro-geologically connected to CSA. It is possible for wells to be contaminated by up-gradient sources of VOCs, including TCE, that migrate in the groundwater layer itself; thus, wells #3 and 4 may be vulnerable to contaminants originating from BP.

Numerous toxic chemicals have been identified in soils, surface water, sediments, and groundwater [70–78]. In the 1980s and 1990s, mandated testing detected chromium levels above the MCL. Further, a MCP Phase II Risk Assessment in 1994 detected levels of acetone, benzene, PCE, TCE, toluene, and cadmium in soil and/or groundwater above MCLs. Moderate to high TCE has been detected in every on-site test performed on shallow groundwater. High concentrations of chemicals including TCE and its degradation product 1,2-DCE were also measured in neighboring private drinking wells [71]. Although the Bird Property is not deemed a significant risk to human health because the localized BP groundwater is not an active drinking water source, models conducted by MDEP and others have not explored the plausibility of toxic contaminants migrating off-site and down-gradient towards the supply wells and into the adjacent Cedar Swamp recharge area/Zone II for Wells #3, #4 (Figure 1) This is another example of aquifer vulnerability not being adequately gauged with respect to runoff/wastewater flows from a waste site to well capture zones and/or aquifer recharge zones (Figure 1). In SWAP, lack of significant risk is based on the absence of designated “wastewater” flows to surface water sources (not present in Holliston); such criteria are inadequately protective of groundwater sources/aquifers.

According to diagrams created during the 2005 Phase II assessment, groundwater contamination plumes are moving in the direction of predominant groundwater flow (northwest-to-southeast, Figure 1), across Marshall Street and directly into Cedar Swamp. Notably, the 1994 Phase II Risk Assessment of BP found site hazard indices (HI) between 5.2 and 48.3 based on groundwater sampling, where $HI > 1.0$ represents an unacceptable additive non-cancer risk assuming use of the water for

drinking and bathing [78]. Total site cancer risk estimated for hypothetical consumers exceeded the Cumulative Cancer Risk Limit of 1.0×10^{-5} .

Testing conducted within Cedar Swamp has been minimal. Only one surface water sample is recorded from 1987 testing [71,77] and only one location (30 Marshall St.) has been tested since 1987. The extent of the groundwater plume off-site is thus unknown. Cleanup activities were conducted between 1987 and 2005, but a finding of “no significant risk” has yet to be accepted for this site. Nevertheless, a proposed low- to middle-income housing development has recently been approved, overturning previous denials based on pollution concerns.

4.1.5. Lake Winthrop

Lake Winthrop (LW) is a recreational lake used for fishing, swimming and boating. The pond suffers from nutrient loading by fertilizer run-off from adjacent residential lots, and is also susceptible to microbial contamination from septic systems. Until the 1970s, herbicides (Silvex, Kuron or 2,4,5-T) were used to control weeds choking the 100-acre pond. In 1984, fish and sediment from Lake Winthrop were collected by the state as part of the Six Ponds Dioxin Survey and analyzed for 2,3,7,8-TCDD, a by-product of the applied herbicides [79]. Yellow and brown bullhead fish ($n = 5$; 2–4 years old; 227–283 g; 255–302 mm in length) and vertical, cylindrical core samples of bottom sediment ($n = 2$) were collected. TCDD concentrations for composite fish and sediment samples were 71 and 5.9 pg/g, respectively, exceeding the USDA guideline of 50 pg/g in fish flesh for human consumption. In 1996, additional fish samples ($n = 9$) were collected: three largemouth bass, three yellow perch, and three yellow bullhead. The average (SD) TCDD concentrations (pg/g) for species composites ranged from 0.12 (0.18) to 0.42 (0.47). In the Survey, there is no mention of water samples being taken from the lake or from Well #1, located on the shoreline (Figure 1).

Between 1996 and 1997, fish were again sampled for metals, dioxin, organochlorine pesticides (OCPs) and PCBs. Samples included largemouth bass ($n = 3$), yellow perch ($n = 3$), bluegill ($n = 2$), pumpkinseed ($n = 1$), and yellow bullhead ($n = 3$). Mercury concentrations for species composite samples ranged from 0.29 to 0.70 mg Hg/kg wet weight, exceeding the Massachusetts Department of Public Health’s trigger level of 0.5 mg Hg/kg for the composite of largemouth bass and yellow bullhead. OCP and PCB levels were below detection limits [80–82]. TCDD levels ranged from 0.2 to 0.4 pg/g and were lower than in 1984. Most recently, however, dioxin levels from sediment sampling in 2012 were higher than those reported in 1983 and above MCP guideline levels [83]. Signs warning the public against fish consumption are posted at beaches and boat ramps.

4.2. Contaminant Levels in the Aquifer

Secondary data on contaminant concentrations in aquifer water were compiled from quarterly SDWA-mandated sampling of drinking water at 5 to 6 sites (see Figure 1 for monitoring sites), and from waste site data. Although limited in spatial and temporal resolution, these data show exceedances of MCLs (Table 3). For example, TCA and TCE were detected at high levels in groundwater at AC site (100–1000 times higher than MCLs). Mean drinking water levels of Mn were four times the U.S. EPA infant health advisory at Well #6; and Hg levels in surface water adjacent to CRC were more than 1000 times the MCL in 2005.

Table 3. Levels of Mn, Hg, trichloroethane (TCA), and trichloroethylene (TCE) from secondary data.

| Chemical | Concentrations (mg/L) | Location Detected | Date |
|-----------|---|-------------------------------|------------------|
| Mn | Mean (SD): 1.25 (0.81), range: 0.0–3.4 (vs. secondary MCL 0.05, infant HA 0.3) | Well #6 | 1987–2013 |
| Mn | Mean (SD): 0.74 (1.08), range 0.0–3.8 | CRC monitoring wells | 2005–2008 |
| Hg | 9.10 (vs. MCL 0.002) | CRC site, adjacent wetland | 6 September 2005 |
| 1,1,1-TCA | 19.3 (vs. MCL 0.20) | monitoring well #5 at AC Site | 17 March 2006 |
| TCE | 8.99 (vs. MCL 0.005) | monitoring well #5 at AC Site | 17 March 2006 |

4.3. Contaminants of Interest

The site profiles detailed above, together with secondary data on chemicals detected in aquifer water in the area were used to generate a list of contaminants of interest (Table 4). These contaminants were detected more frequently in environmental samples, are held to National Primary Drinking Water Regulation (NPDWR) standards, and/or have been associated with adverse health effects in epidemiologic studies.

Table 4. Contaminants of interest identified from site profiles and secondary data.

| Chemical Class | Contaminants of Interest |
|----------------------------------|---|
| Metals (5) | Cadmium, Chromium, Lead, Manganese, Mercury/Methylmercury |
| Chlorinated solvents (6) | 1,2-Dichloroethylene (DCE), 1,2-Dichloroethane (DCA), Dichloromethane (DCM), Trichloroethylene (TCE), 1,1,1-Trichloroethane (TCA), Tetrachloroethylene (PERC) |
| Trihalomethanes (THMs) (3) | Chloroform, Bromodichloromethane, Chlorodibromomethane |
| Flame retardants | Polybrominateddiphenylethers (PBDEs) |
| Polyaromatic hydrocarbons (PAHs) | Benzo-a-pyrene |
| Other combustion hydrocarbons | 2-Methylnaphthalene, Benzene/Toluene/Ethylene/Xylene (BTEX) |
| Radionuclides | Gross beta particles |
| PCBs | PCB congener 1242 |
| Dioxins | 2,3,7,8-TCDD |

4.4. Aquifer Vulnerability

The hydrogeological characteristics of the two local aquifers (BBA, CSA) used as drinking water sources in Holliston make them particularly vulnerable to contamination from anthropogenic sources, such as spills or underground discharges of wastes. The aquifers comprise highly permeable sand and gravel, and the aquifer layer is very shallow (<20 feet to water table, <50 feet to bedrock) [19]. Furthermore, there are no natural clay barriers to confine the movement of contaminants horizontally. Importantly, the aquifers are also vulnerable to persistent contamination from naturally occurring Mn in Dedham Granite bedrock.

Table 5 summarizes our rating of Holliston aquifer vulnerability. The overall rating of 0.88 indicates high vulnerability. Simple ratings allow aquifers to be mapped and compared in geographical regions like New England, as a way to inform integrated regional assessments. GIS-based SWAP would allow local, regional and national scales to be considered *concurrently* to inform strategic actions, in partnership with U.S. EPA and U.S. Geological Service.

The 2002 Holliston SWAP report recommended that Zone I for all wells be set at a 400-foot radius. It further states: “The Zone II for Well #5 and Well #6 contains DEP Tier Classified Oil and/or Hazardous Material Release Sites indicated on the map as Release Tracking Numbers 2-11053 2-0063, and 2-0065. Currently, the Town does not have water supply protection controls that meet DEP’s Wellhead Protection regulations 310 CMR 22.21(2). Protection planning protects drinking water by managing the land area that supplies water to a well. A Wellhead Protection Plan coordinates community efforts, identifies protection strategies, establishes a timeframe for implementation, and provides a forum for public participation.” (p. 7 [18]). The Release Tracking Numbers, however, belong to three other sites (one with TCE contamination, two with oil), but not to WTS or CRC. Consistent with our findings, two wells assessed under SWAP are of moderate susceptibility (#1, 2), while four are high (#3, 4, 5, 6).

Table 5. Holliston aquifer vulnerability rating.

| Criterion | Holliston Aquifer Descriptor | Vulnerability Rating * |
|---|--|------------------------|
| Depth to water table (unconfined aquifers). | Very shallow (0–20 feet). Low-lying areas where wells sited often swampy. | 3 |
| Depth to bedrock/source of natural contaminants (e.g., Mn). Source strength. | Shallow (30–60 feet), fractured granite source of Mn. Moderate strength (based on limited Mn data). | 2 |
| Aquifer material. Presence/absence of natural clay barriers to contaminant dispersion/transport. | Unconsolidated sand-gravel, glacial till. No natural clay barriers present. | 3 |
| Topography, stratigraphy and hydrologic sensitivity. | Surface elevation difference between hills and valley about 300 feet, gradients quite steep. Aquifer and Mn levels likely to be highly sensitive to precipitation events. Mn variability likely high esp. temporal. | 2 |
| Reliance of population on shallow aquifer. | 100%—about 14,000 consumers. Private wells were discontinued because of TCE levels down-gradient of/proximate to Bird Property. | 3 |
| Treatment of source water. | Limited, not on all wells—installed systems have trouble coping with Mn levels. | 2 |
| Reliance on septic systems/microbial risk. | 100% of homes, businesses rely on septic and leach fields, incl. new high-end homes on hills. | 3 |
| Sources of anthropogenic contaminants (esp. toxics, hazardous waste sites). Type and number of contaminants, source strengths vary. | Several sources, sites of concern. From profiling four, and data from fifth, there many contaminants of interest (Table 4), but source strengths are uncertain. Two are existing, operational form many years. Others legacy, including US Superfund sites and Mass DEP 21E sites. | 2.5 |
| Monitoring of drinking water quality, and effectiveness of regulation. | Limited temporal and spatial monitoring: one sample every 3 months at 6–8 sites. Monitoring is the minimum effort required under the SDWA and Mass. State law. | 3 |
| Responsiveness of water agency, local policy makers. | Local Selectmen and Administrator have taken a defensive stance, claiming water is safe. | 3 |
| Presence/Absence of a Comprehensive Wellhead Protection Plan. | Recommended by 2002 SWAP, but none seems to have been developed | 3 |
| Awareness level among residents of potential risks. | Mixed awareness, ranging from some very concerned residents and others with no concerns. Energy among those concerned is high. | 2 |
| <i>Overall rating (31.5/36)</i> | | <i>0.88</i> |

Notes: * Vulnerability: 1—low; 2—moderate; 3—high.

4.5. Well #6 Capture Zone and Well Service Areas

The preliminary analytical model estimates a mean capture-zone width (± 1 SD due to variable pumping rate) of 2300 (± 780) feet for well #6 at the up-gradient location of the CRC (Figure 3). The estimate from the Visual AEM model is comparable. These findings corroborate that CRC is located within the SWAP Zone II buffer. The CRC is adjacent to the Zone I buffer set at 400 foot radius; yet, surprisingly there is no specific mention of CRC in the SWAP Report [18]. This finding has major implications for regulation and permitting under the SDWA/SWAP. The WTS is on the edge of well #6's Zone II, allowing its wastewater and runoff flows to potentially compromise the capture zone of well #6, and possibly well #5, which is a high-pumping well down-gradient of CRC and WTS (Figure 3).

Based on our model of service areas for each well, we estimated that each home receives its drinking water from a primary well, but many are also able to receive water from a secondary well if the primary well is down for maintenance. This finding is consistent with engineering design and hydraulics, but runs counter to the perception/official claim that residents always obtain water from a mixture of wells. The estimated service areas for wells #1–6 ranged from 3300 feet to 11,500 feet in radius from the well, providing coverage and some redundancy as expected (Figure 4).

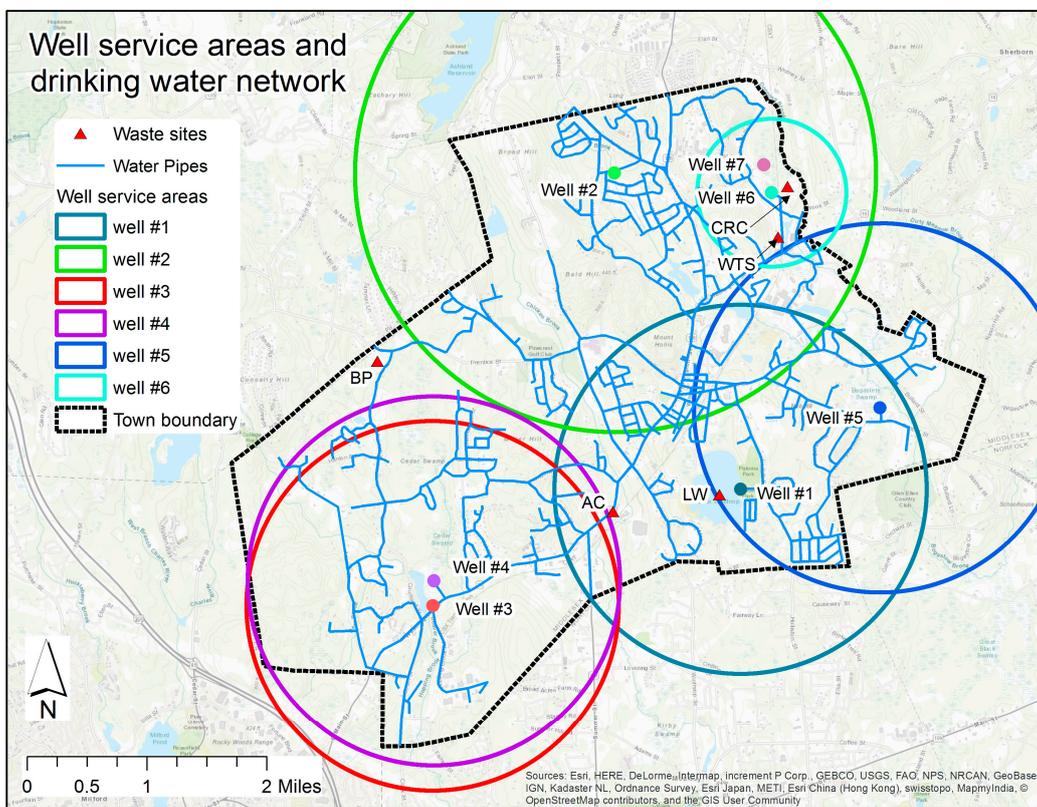


Figure 4. Drinking water network and estimated well services areas in Holliston. The service areas are homes served by the pipes that lie within each circle: the total dynamic head (TDH) that the reference pump can achieve is calculated based on the actual pipe network that lies within the circle. Note: The service areas for Well #2 and #6 do not overlap; #2 is able to serve low-elevation areas within the green circle only, while #6 serves high-elevation areas. The small area outside the circles (i.e., around BP) suggests that the model slightly underestimates service areas for Wells #2 and/or #4. Data sources: Bureau of Geographic Information (MassGIS), Commonwealth of Massachusetts, Executive Office of Technology and Security Services, Holliston Water Department.

5. Recommendations

Several recommendations for scientific research, policy and regulation, and technology arise from this case:

1. Environmental policy, regulation and practice:
 - Utilize an integrative approach to the regulation of multiple sites, contaminants and exposures in order to increase the likelihood that environmental resources are adequately protected from harmful pollution.
 - Improve permitting/siting of waste-generating facilities to help ensure that facilities and their runoff and/or groundwater plumes cannot impact well buffer Zones I and II.
 - Increase protection of buffer zones: Strengthen SWAP in its setting and ongoing evaluation of buffers to protect shallow aquifers. Improve modeling and delineation of buffer protection zones, in particular Zone II.
 - Utilize a systems-based integrated assessment rather than a conventional site-by-site/spill-by-spill risk assessment approach, including GIS to combine and analyze data.
 - Strengthen drinking water monitoring protocols in order to capture spatio-temporal water quality variability, with extended sampling beyond the currently limited temporal (quarterly) and spatial resolution (5–8 sites). In Consumer Confidence Reports (CCRs), improve upon reporting of variability in exposures, as reporting is currently limited to global mean values.
2. Public health policy, regulation and practice:
 - Utilize an integrative approach to the assessment and management of multiple exposures to more accurately estimate health risks and inform cost-effective risk mitigation.
3. Environmental technology:
 - Develop new water treatment technologies to remove Mn from drinking water, given frequent detection of naturally occurring Mn in aquifers in the U.S. [84] and the inability of existing water treatment plants to adequately remove Mn (e.g., in Holliston).

6. Conclusions

Complex exposures to multiple contaminants in air, water and food were recently estimated to account for one-in-six deaths globally in 2015 [85]; Holliston illustrates this complexity on a local scale. Our integrated assessment reveals moderate to high levels of Mn, Hg, and solvents detected in well water, soil water or groundwater at multiple locations from the 1960s to present. Our qualitative analysis of aquifer vulnerability used a rating scheme that considers geophysics, hydrogeology, contaminant chemistry, regulation and social awareness, and may be applied to other aquifers. The overall vulnerability rating suggests a highly vulnerable aquifer system. Based on modeling one well's capture zone and residential service area, it is likely that multiple chemicals entered the drinking water supply, exposing residents.

Importantly, the five criteria necessary for an exposure pathway to be considered complete are present in Holliston: (i) source(s) of contamination include the five waste sites profiled above; (ii) the environmental media and transport mechanisms are groundwater flows and surface runoff that impact aquifers; (iii) point of exposure is at the household tap; (iv) route(s) of exposure include drinking, eating, and dermal contact; and (v) the population of Holliston utilizing the public drinking water represents the receptor population [43]. Based on the weight of evidence, a complete pathway to exposure for several chemicals of concern is likely in Holliston. Although a complete exposure pathway does not necessarily mean that a public health hazard exists, it does warrant closer examination.

We identified several contaminants of public health concern, including Mn, Hg, and TCE. Mn is an essential nutrient but also a neurotoxicant at excess levels. Few studies have evaluated links between

Mn levels commonly found in U.S. drinking water aquifers and neurobehavioral outcomes [86,87]. Exposure to all forms of Hg are known to adversely affect the developing fetus, and there is no beneficial role of Hg in the human body [88]. Solvents such as TCE are common aquifer contaminants and exposure to TCE has been linked with behavioral alterations in animals [89,90]; yet, few human studies have examined the neurotoxic consequences of developmental exposures. Individually, these chemicals pose health risks, and in combination, risks may be compounded.

Previous work has examined approaches to groundwater protection policy, including in complex settings of multiple stakeholders with varied priorities [91,92]; however, environmental contaminants that present human health risks are not typically emphasized in those analyses. Other studies have reported on the vulnerability of aquifers to contaminants such as nitrates and pesticides in the U.S. and elsewhere, and these locations may similarly benefit from an integrated assessment that includes an evaluation of exposure pathways and health risks [93–95].

Effective, integrated approaches to tackle real-world environmental health-risk complexity are rooted in vibrant multi-directional exchanges among researchers, affected communities, and policy makers. Such approaches are being encouraged by a growing number of agencies: the findings in Holliston have national and global significance for aquifer protection and human health.

Supplementary Materials: The following are available online at www.mdpi.com/2073-4441/10/01/23/s1, which refers to [23].

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