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Presence and Potential Sources of Pharmaceutical and Personal Care Product Chemicals in Messalonskee Lake

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ABSTRACT

Pharmaceuticals and personal care product chemicals (PPCPs) represent a large group of contaminants that are mostly not regulated in surface or drinking water and whose presence and environmental and health impacts are poorly understood. We investigated the presence and potential sources of 18 PPCPs in Messalonskee Lake, ME. We collected samples four times over the summer of 2017 at 13 sites around the lake at sites of varying human impact. Samples were filtered using solid phase extraction and tested for PPCPs using liquid chromatography-tandem mass spectrometry. We detected five out of the 18 PPCPs we tested for at the ng/L level, including caffeine, 1,7-dimethylxanthine (a caffeine metabolite), acetaminophen, sulfachloropyridazine (a veterinary antibiotic), and amphetamine. We saw more PPCP detections in the beginning of the summer (6/26/2017) than at the end of the summer (8/7/2017), although this relationship was not statistically significant. PPCPs showed a significant positive correlation with total nitrogen. PPCPs were more prevalent at public high-use sites like the Oakland public beach and the Belgrade boat launch. Direct human input (urine, dumping, contact) may be one of the major sources of PPCPs in Messalonskee Lake. The significance of septic systems as a source of PPCPs in the lake is still undetermined. The number of detections and concentrations seen in Messalonskee Lake are similar to the other studied lakes that are not impacted by wastewater treatment plants. We also conducted a survey of residents in the Messalonskee watershed to gather information on septic systems and drinking water. Of 33 survey respondents, 25 reported having their septic system pumped within the past three years, 16 reported having had their septic system inspected within the past three years, and 18 reported having a septic system under 20 years old. Only one survey respondent reported taking their drinking water directly from the lake. Messalonskee Lake is not highly contaminated with PPCPs, but the potential ecological and human health impacts of long-term exposure to the low levels of PPCPs found in this study should be investigated.

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INTRODUCTION

Maine's lakes are an indispensable resource in terms of ecological value, tourism and recreation, and property values, generating 3.5 billion dollars annually. Additionally, for nearly half of Maine's population that rely on public water supply, these lakes provide a source of drinking water (ME DEP n.d.). It is therefore vitally important to maintain these waters and to protect them from contamination.

There are about 400 thousand hectares of lakes in Maine, which accounts for 4.5% of the state's total area. Since the 1950s, Maine has classified its surface waters in order to establish goals for the maintenance and improvement of water quality across the state (Bacon 2013). This initiative requires the Maine Department of Environmental Protection to evaluate all surface waters at least every three years, which has led to the close monitoring and improvement of many waters across the state and to the reputable water quality that Maine boasts today (Meidel 2016). All lake water must be of "suitable" quality for drinking after disinfection, along with other requirements such as being suitable for recreation and fishing (Maine State Legislature 2013; Bacon 2013).

However, it is important to note that this program focuses largely on water quality in terms of nutrient overloading from sewage effluent (Meidel 2016). While this issue is indeed of great concern for ecosystem health, human health, and property value, there are currently no regulations regarding the quality of surface waters in terms of another class of pollutants called emerging contaminants (Richmond et al. 2017; Tarpani and Azapagic 2018).

Emerging contaminants are synthetic or naturally-occurring chemicals that are present in the environment, notably in surface waters, due to human activities. Within the classification of emerging contaminants are pharmaceuticals and personal care product chemicals (PPCPs), a large subgroup of thousands of chemicals. PPCPs are defined broadly as prescription medicines, over-the-counter drugs, illicit drugs, veterinary drugs and feed additives, certain cosmetics, and certain foods/food supplements, along with their respective metabolites and transformation products (WHO 2012).

PPCPs form a large class of emerging contaminants, which means that they are not officially monitored or reported by any agency regardless of whether they are present as

pollutants in wastewater effluent or even in drinking water (Boles and Wells 2010). They also do not fall within protections of the Clean Water Act or Safe Drinking Water Act (Wilson et al. 2003). PPCPs are a group of products that continues to grow as the Food and Drug Administration approves the distribution of over 20 substances per year (Pease et al. 2017).

PPCPs are not commonly monitored but may present risks to humans and/or the environment (Kolpin 2017). Recently, the US Geological Survey (USGS) has been tasked with investigating emerging contaminants in order to understand their occurrence in the environment, their sources, their transport and fate within the environment, and the ecological effects of their presence (Kolpin 2017). The USGS has described the complex mixtures of PPCPs in streams and has investigated ecological impacts in certain aquatic organisms (Kolpin 2017). Despite the USGS mandate, many surface waters have not been assessed, including most lakes.

Since lakes serve as a primary source of drinking water for two-thirds of Maine, it is important to also determine the potential impacts on human health of low-level, long-term exposure to PPCPs. Studies in Minnesota have identified the presence of PPCPs in lakes that range from pristine and completely undeveloped to highly impacted and urban (Ferrey 2013; Ferrey 2015). Their findings have shown that less developed lakes tend to have fewer contaminants present, but they have also shown that with very few exceptions even the most pristine, remote lakes show detectable amounts of some emerging contaminants, like DEET, cotinine, and BPA (Ferrey 2015).

A key first step is to determine which chemicals are present and at what levels. Secondly, determining sources of emerging contaminants is important in order to pinpoint areas where improvements may prevent contamination in the first place. Past studies have focused particularly on determining the removal rates of emerging contaminants at wastewater treatment plants (WWTPs) and at drinking water treatment plants (DWTPs; Deo 2014; Yang et al. 2017). Sources of emerging contaminants in surface water include WWTP effluent, failing septic systems, and direct humans uses, notably during recreational activities (Petrović et al. 2003).

Wastewater Treatment

Incomplete wastewater treatment may be the most widespread and significant form of environmental PPCP contamination (Petrie et al. 2015). Municipal WWTPs, private septic systems, and cesspools all contribute to PPCPs polluting surface and ground waters. WWTPs represent the highest aquatic loads of PPCPs and are the most easily traceable source (Petrie et al. 2015).

Wastewater Treatment Plants

Municipal sewage treatment is the most widely-used form of wastewater disposal in the U.S, serving over 80% of the population (Petrie et al. 2015; Center for Sustainable Systems, University of Michigan 2016). The current most common method of wastewater treatment involves separate sanitary sewers, which are designed to only collect wastewater and typically only allow the entry of storm water through cracks and leaks. This means that the potential for sewer overflow is relatively low when systems are in good condition (EPA 2016a).

The U.S. Environmental Protection Agency (EPA) requires public WWTPs to uphold secondary treatment processes, which include minimum technology-based requirements that aim to achieve standards for five-day biochemical oxygen demand, total suspended solids removal, and pH (EPA 2016b; Figure 1). Sometimes, when effluent-receiving waters require higher water quality, sewage must undergo processes of tertiary treatment, which basically seek to further reduce suspended solids (and therefore biological oxygen demand), control pathogenic viruses and microorganisms, remove excess nutrients, and remove trace elements and organic compounds (National Research Council 1996).

Depending on the different levels of processing and treatment efficiency through which wastewater goes, the resulting effluent may cause varying levels of contamination to receiving surface waters (Petrie et al. 2015; Kolpin et al. 2002). Because WWTPs are not required to remove any amount of PPCPs from wastewater during the treatment process, many PPCPs remain dissolved or suspended in the effluent, while some substances are coincidentally effectively eliminated through photolysis, adsorption, biodegradation (Lin et

al. 2010; Bagnall et al. 2013). Whether PPCPs adsorb to sewage sludge and get removed from the wastewater depends on their hydrophobic and electrostatic properties.

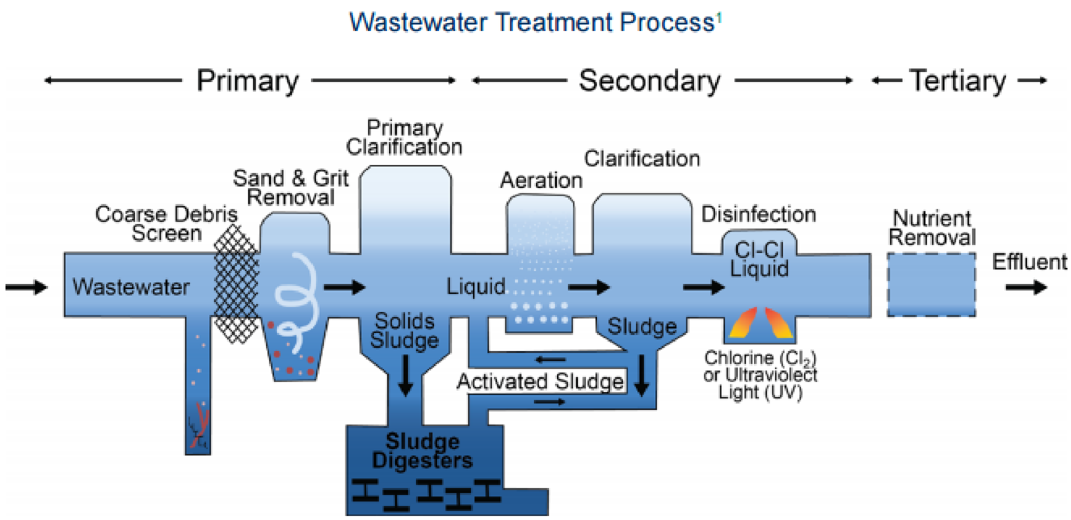

Figure 1. A diagram of the municipal wastewater treatment process, including the primary, secondary, and tertiary processes. (Center for Sustainable Systems, University of Michigan 2016).

Septic Systems

Importantly, half of Maine's households use a septic system, and at the state level there are no regulations to ensure proper maintenance of these systems once they are first installed and inspected. This means that septic system leakage may be a major contributor to emerging contaminant pollution in Maine.

Septic systems, also called onsite wastewater treatment systems, are a common method of wastewater treatment in which private homeowners are responsible for the maintenance of the system. Septic systems are used by between 25 and 30% of rural and waterfront households in the US (Verstraeten et al. 2004), meaning that the issues associated with septic system leakage and environmental contamination are significant. The potential

for septic system leakage of PPCPs is especially relevant to this study, as the vast majority of Messalonskee Lake watershed residents use septic systems for their wastewater treatment.

There are five major components to septic systems, including a pipe from the home that transports the wastewater, the septic tank, the drainfield, and the soil. The septic tank is typically made of concrete, fiberglass, or polyethylene, and is an underground watertight container that holds wastewater and allows time for solids to settle to the bottom (sludge) and to the surface (scum) while also partially treating wastewater through bacterial digestion. Wastewater is then pushed out to the drainfield as new wastewater enters the system, and the partially treated water continues to be treated by percolating through the soil, which aids in removing harmful bacteria, viruses, and nutrients before the wastewater finally reaches groundwater (EPA 2005).

Ensuring the proper functioning and maintenance of a septic system requires that it be inspected every one to three years and pumped every three to five years (National Environmental Service Center 2004; EPA 2017). However, in many towns there are no laws forcing homeowners to regularly inspect or pump their septic systems. This means that many septic systems go unmaintained, resulting in significant environmental contamination (Withers et al. 2014).

There are two types of septic system failure: hydraulic failure and phosphorus treatment failure. Hydraulic failure occurs when sewage over flows due to too much wastewater in the soil. Phosphorus treatment failure, on the other hand, happens when soils are ineffective at removing the excess nutrients and other substances from the wastewater and can occur even when the system is properly maintained (O'Hara n.d.). It is even possible for properly maintained septic systems to leach excess nutrients and PPCPs into groundwater, contaminating well water (Kozuskanich et al. 2014). A study of septic system leakage in soil samples on East Pond (another of the Belgrade Lakes), concluded that septic systems are not likely a significant source of contamination in the lake (Reed and Haver 2015).

Drinking Water

52% of Maine's population relies on private drinking water from wells or lakes, and the maintenance of private drinking water, including from private wells, is unregulated in Maine (ME DEP 2016), meaning that people may employ a wide variety of filtering systems that remove varying levels and types of emerging contaminants from their water and may not recognize PPCPs as a potential issue.

Residents of Kennebec County obtain their drinking water from a variety of sources, due especially to the fact that the county represents a diverse mix of urban and rural areas. The conventional treatment process of public water supply typically consists of four steps, including coagulation and flocculation to create larger particles; sedimentation to allow the larger particles to settle out; filtration through sand, gravel, and charcoal filters to remove smaller dissolved particles such as dust, parasites, bacteria, viruses, and chemicals; and lastly, disinfection by the addition of chlorine or chloramine (or sometimes ozonation) in order to disinfect the water, especially as it is transported through pipes (CDC 2015). Some PPCPs are effectively removed by the conventional drinking water treatment process (amphetamine-type stimulants (99%), cocaine (100%), nicotine (100%), sulfamethoxazole (100%)) while other substances, such as caffeine (90%), carbamazepine (85%), and cotinine (74%) are only partially removed (Huerta-Fontela et al. 2008; Stackelberg et al. 2007). While municipal water usually complies with EPA standards, PPCPs may still be in public drinking water, unmonitored and unreported.

People obtaining their drinking water privately are not federally required to uphold the standards set by the Safe Drinking Water Act, and only some states and towns require regular inspections. Methods of privately sourcing water include drilled wells, driven wells, dug wells, and lake water. Approximately 15% of the US population drinks well water. Dug wells, usually around three to nine meters deep, are the shallowest type of well and are also the most likely to be contaminated. Driven wells are also relatively shallow, at around nine to 15 meters deep, and have a medium to high risk of contamination depending on the exact depth and on the nearby land-use activity. Drilled wells, at 30 to 120 meters deep, are the safest type of well in terms of contamination risk. Septic systems and their leach fields have

the potential to put wells at risk, an unfortunate relationship given that houses that use well water are also more likely to have septic systems (EPA 2016c). Private, drilled wells are the most common source of drinking water in the Messalonskee watershed.

People living adjacent to surface waters may in some cases choose to obtain their drinking water by piping it directly into their houses, especially if they are seasonal residents. Lake water is rarely of high enough quality for drinking, as it is highly affected by runoff, animal excretions, and pollution from boaters and machinery. Individual filtration systems may improve the water quality enough for it to be a practical water source (Fleck, n.d.). Filtration systems for lake water are even less regulated than for private wells, however, meaning that people who obtain water in this way may not follow the procedures necessary to ensure their health. Given the tradeoffs between privately sourcing drinking water from well versus surface water, the population around Messalonskee Lake do both. Unfortunately, there are no public records regarding which houses get their drinking water from which source.

Pharmaceuticals and Personal Care Product Chemicals

PPCP Sources

PPCPs are released into the environment largely through sewage effluent, the treatment of which is often unequipped and ineffective at removing many of these compounds (Petrović et al. 2003; National Environmental Service Center 2007; Petrie et al. 2015; Figure 2). However, there are other significant sources of PPCPs in the environment that must be considered in order to understand their pervasiveness. Septic systems, for example, become problematic when there is leakage, often a result of a lack of proper monitoring and maintenance by property owners (Fairbairn et al. 2016). PPCPs can also be released into the environment directly through agricultural and animal farming runoff, landfills, dumping and through urination or direct contact between skin and surface waters (Petrović et al. 2003). PPCPs have even been detected in supposedly pristine lakes, suggesting that they can also be carried as particles through the air and deposited with rain, therefore able to contaminate surface waters to a certain extent regardless of the level of development or direct inputs into the water (Ferrey 2013).

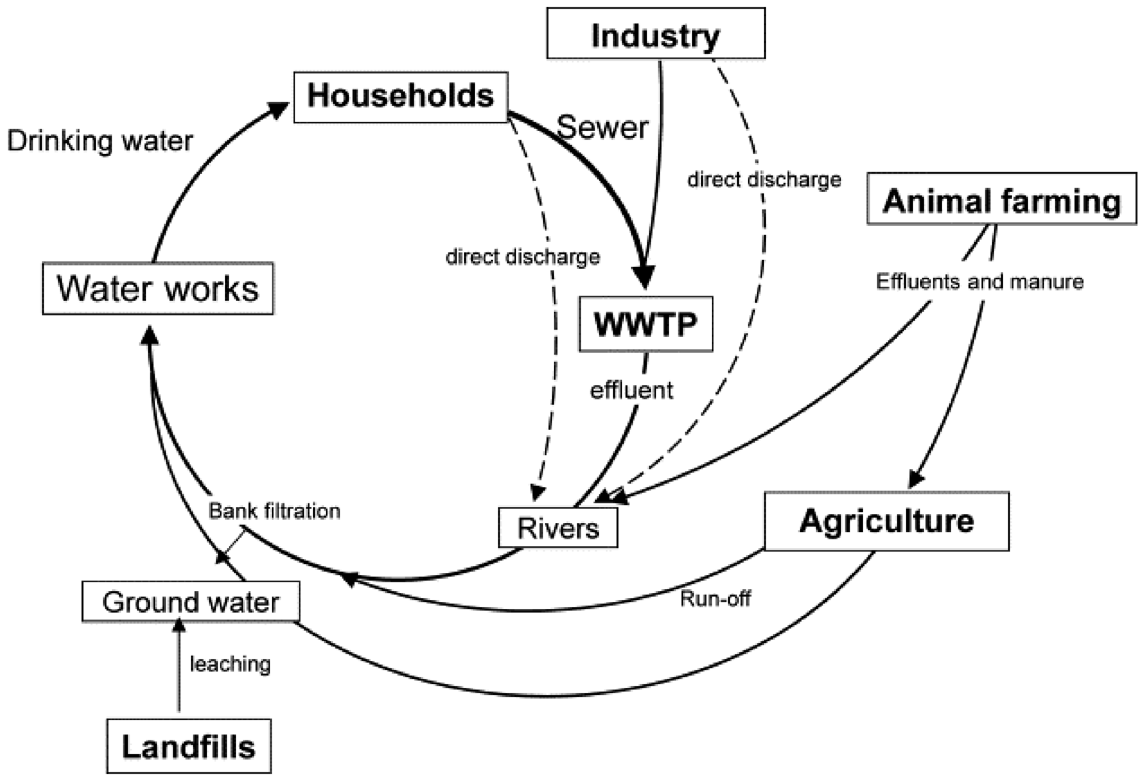

Figure 2. Transport of PPCPs through the environment (from Petrović et al. 2003).

There is evidence that some PPCPs fluctuate temporally in the environment for a variety of reasons. PPCP concentrations tend to be higher both when water conditions are cold and flow is low due to reduced degradation in the environment and also when water is warm and the flow rate is high due to increased flow through WWTPs with reduced retention time, lowering the removal efficiency of many PPCPs (Kolpin et al. 2004; Fairbairn et al. 2016). Other studies have found increased PPCP concentrations during winter months and decreased levels in the spring and summer (Vieno et al. 2005; Veach and Bernot 2011; Huerta-Fontela et al. 2008). Huerta-Fontela et al. (2008) also found weekly fluctuations in the levels of certain substances, like the concentrations of some controlled and illicit drugs, which were found to be higher from Friday to Monday. Agricultural herbicides tend to spike in concentration early in the summer when application rates are the highest and higher levels of precipitation increase runoff to surface waters (Fairbairn et al. 2016).

Many PPCPs are effectively degraded in relatively short periods of time (from a few hours to a few weeks) through processes such as photolysis, biodegradation, and sorption into sediments (Table 1). However, their often-continuous introduction into the environment causes them to present characteristics and effects of persistent chemicals (Figure 1; Petrović et al. 2003). It is also important to note that the degradation characteristics of many PPCPs are not yet known and can depend on often vastly different environmental conditions (Wilson et al. 2003).

PPCP Impacts on Human Health

The human health impacts of continuous, long-term exposure to PPCPs in drinking water and in the environment remain poorly understood (Kolpin et al. 2002; Zuccato and Castiglioni 2009). Official standards for contaminants in drinking water are provided by WHO and the EPA for a limited number of PPCPs (WHO 2012). A case study on the Rhine and Meuse Rivers in Europe showed that for the majority of PPCPs for which there is either an EPA or World Health Organization guideline, there is a substantial margin of safety between the guideline and the maximum concentration encountered (Schriks et al. 2010). This suggests that many PPCPs individually may not pose a significant threat to human health. However, this represents only one case study, and many PPCPs remain unevaluated and without standard guidelines to follow. This does not take into account aggregate exposures to many PPCPs, many of which may have similar health impacts, or long-term exposures. Also, the health guidelines may not consider vulnerable populations.

Additionally, it is possible that PPCPs may form complex mixtures and create cumulative and synergistic effects even at low concentrations (Kolpin et al. 2002; Petrie et al. 2015). Such synergistic interactions between PPCPs at environmentally-realistic levels have been shown experimentally to inhibit human embryonic cell growth by between 10 and 30% (Pomati et al. 2006), and different mixtures and levels of exposure can change the observed effects on cells (Pomati et al. 2007).

Understanding of ecological impacts of PPCP contamination in the environment is likewise limited despite two decades of research. This is probably due to the diverse array of contaminants classified as PPCPs and also to the distinct ecosystems with which they interact

(Boxall et al. 2012). Schnell et al. (2009) found that in rainbow trout, the toxicity of certain PPCPs in combination is additive and can occur at lower-than-expected concentrations due to synergistic effects. Since the combinations and levels of PPCPs will be very different on both small and large spatial scales, predicting these synergistic impacts will be difficult.

Since it is not feasible to test for the presence of every PPCP, many studies have focused on a subset of PPCPs with similar enough chemical properties to simplify analytical methods (Kolpin et al. 2002). This study was limited to the analysis of 18 PPCPs commonly found in surface waters which could all be tested for using liquid chromatography-tandem mass spectrometry and positive and negative ion detection methods (Water Sciences Laboratory, Lincoln, NE). The suite of PPCPs included in this study are three stimulants and stimulant metabolites, three over-the-counter medications, six prescription medications, three recreational drugs, and three agricultural/veterinary compounds (Table 1).

Research Question and Goals

The primary goal of this study was to determine the presence or absence and concentration of 18 PPCPs in Messalonskee Lake (Snow Pond) in Kennebec County, ME. The second major goal of this study was to identify possible sources of any PPCPs that were detected, specifically in relatively low-impact surface water. The final goal of the study was to explore whether there might be possible health risks to people living around Messalonskee Lake due to direct exposure to PPCPs because of lake water being used as a source of drinking water.

Messalonskee Lake is different from many other surface waters that have been tested for PPCPs in that it is not located downstream of a WWTP. This is one of only a handful of studies that have focused on relatively low-impact surface waters to explore the role of PPCP sources other than WWTPs (Ferrey 2013; Ferrey 2015; Haver 2016). Because there are no WWTPs on the Belgrade Lakes it may be more pristine than other lakes, but there is considerable residential development along the lakeshore.

This research builds on the only other known study on PPCP presence in the Belgrade Lakes. In December 2015, samples collected on Great Pond, East Pond, and Long Pond were

found to have d-amphetamine, caffeine, and 1,7-dimethylxanthine (a caffeine metabolite); these chemicals were more likely to be present at higher usage areas on the lakeshores like boat launches (Haver 2016; Figure 3). This was a surprising result because even during a very low-use period of the year PPCPs were detected. Samples were collected at one point in time from each site, meaning that the residence time of present PPCPs could not be estimated.

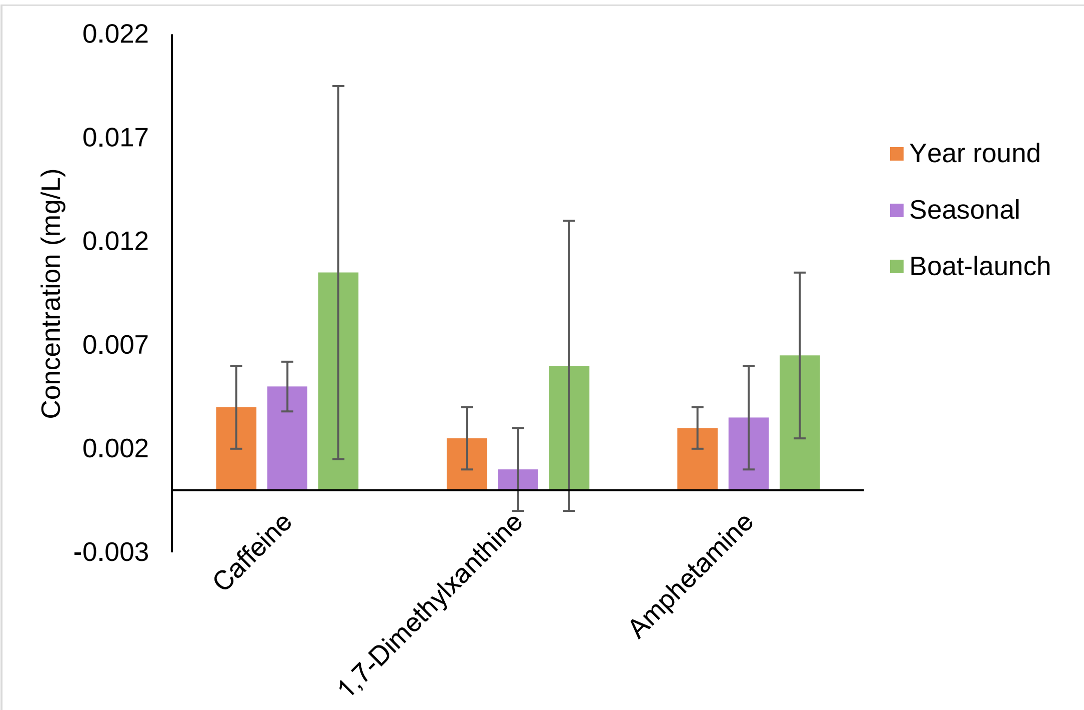

Figure 3. Average concentrations (mg/L, with standard error) of three detected PPCPs in Great Pond, East Pond, and Long Pond in November 2015, grouped by human use at each site (adapted from Haver 2016).

The present study builds on the study by Haver (2016). We collected samples at many sites on one lake to allow for higher spatial resolution on a limited budget. Also, samples were collected four times over six weeks, providing a more detailed view into the residence time of detected PPCPs. Sampling in the summer meant capturing PPCP loads during a more

active period of the year in terms of human activity on the lake (recreation, seasonal residents), when concentrations would supposedly be at their highest. Budget limitations meant we were unable to carry out this analysis on more than one lake.

We also conducted a survey of Messalonskee Watershed residents to (1) gather information about the condition of septic systems in the Messalonskee Watershed, which may be a source of PPCPs, and to (2) collect information on how residents source their drinking water, with attention to the potential public health concerns if residents take their drinking water directly from the lake.

The following sections describe the uses, breakdown methods and rates (Table 1), and ecological and human health impacts of the 18 PPCPs tested for in this study.

Table 1. 18 PPCPs analyzed in this study, their common uses, how they break down in surface waters, and how long it takes for them to break down.

Chemical name	Description	Method of breakdown (half-life in hours)	Reference
STIMULANTS			
Caffeine	Stimulant	Sorption and Biodegradation(36 – 72)	Lin et al. (2010); Pérez et al. (2005)
1,7-dimethylxanthine	Caffeine metabolite	--	--
Cotinine	Nicotine metabolite	--	--
OVER THE COUNTER MEDICATIONS			
Acetaminophen	Pain and fever	Photolysis (34 – 55); Sorption and Biodegradation (26-50)	Yamamoto et al. (2009); Lin et al. (2010)

Cimetidine	Antihistamine, for gastrointestinal disorders	Likely Photooxidation (resistant to photolysis)	Latch et al. 2003
Diphenhydramine	Antihistamine, for minor and severe allergic reactions	Catalyst-aided photolysis	Pastrana-Martínez et al. (2012)
PRESCRIPTION MEDICATIONS			
Amphetamine	Stimulant, for ADD, narcolepsy	Biodegradation (120)	Bagnall et al. (2013)
Carbamazepine	Anticonvulsant, for Seizures, epilepsy	Photolysis (84 – 2100); Biodegradation (1400 – 5600)	Yamamoto et al. (2009); Lam et al. (2004)
Morphine	Narcotic, for severe pain	--	--
Phenazone	NSAID and analgesic, for pain and fever	Biodegradation (480 – 720)	Pieper et al. (2010)
Sulfamethoxazole	Antibiotic	Photolysis (430 – 485); Biodegradation (360 – 720)	Lam et al. (2004); Ryan et al. (2011); Pérez et al. (2005)
Trimethoprim	Antibiotic	Photolysis (137)	Lam et al. (2004)
RECREATIONAL DRUGS			
MDA	Psychoactive drug	--	--
MDMA	Psychoactive drug	--	--
Methamphetamine	Psychoactive drug	Biodegradation (190)	Bagnall et al. (2013)

AGRICULTURAL OR VETERINARY COMPOUNDS			
Sulfamethazine	Antibacterial agent for livestock	Biodegradation (360 – 720)	Pérez et al. (2005)
Sulfachloropyridazine	Antibacterial agent for livestock	--	--
Thiabendazole	Fungicide	--	--

Stimulants and Their Metabolites

Caffeine is one of the three most widely detected PPCPs among all studies on emerging contaminants, along with acetaminophen and cotinine (Kolpin et al. 2002; Hedgespeth et al. 2012). WWTPs may remove up to 70% of caffeine when employing ozone treatment methods, and up to 90% from postchlorination (Huerta-Fontela et al. 2008). Although caffeine is one of the most common PPCPs in surface waters, its potential long-term human health impacts have not been explored, possibly due to its frequent consumption by a large portion of the population. Known ecological impacts include changing activity levels and heightening the startle response in *Rana pipiens* tadpoles (Fraker and Smith 2004). It has also been shown to block adenosine receptors in *Lemna gibba* (duckweed), an important aquatic plant species for its nitrogen-fixing properties (Crane et al. 2006).

1,7-dimethylxanthine, also known as paraxanthine, is a caffeine metabolite. It can be effectively removed by sewage treatment (Huerta-Fontela et al. 2008); however, its residence time and degradation in aquatic systems are poorly known. As 1,7-dimethylxanthine is closely associated with caffeine, it was one of the five most frequently detected substances in a suite of 105 PPCPs in a test on 23 Iowa stream sites (Kolpin et al. 2004).

Cotinine is a metabolite of nicotine and is another of the three most widely detected PPCPs (Kolpin et al. 2002; Hedgespeth et al. 2012). About 10 – 15% of nicotine that is consumed by humans is excreted as cotinine (Lopes et al. 2014). Levels of cotinine in

wastewater are temporally variable, with higher concentrations detected on weekends and holidays (Lopes et al. 2014). Standard wastewater treatment processes may remove 90 - 99% of cotinine, but effluent may contain 15 to a few hundred ng/L cotinine (Buerge et al. 2008). Human health impacts due to exposure to aquatic cotinine are not yet known. The ecotoxicological impacts of these levels of cotinine are also not yet well-understood (Buerge et al. 2008), although it is possible that cotinine has similar effects as nicotine, such as increasing release of Ach, dopamine, and glutamine neurotransmitters in the zooplankton *Daphnia pule* (Crane et al. 2006).

Over-the-Counter Medications

Acetaminophen is a common analgesic and is another of the three most widely detected PPCPs (Kolpin et al. 2002; Hedgespeth et al. 2012; Verlicchi et al. 2012). Depending on the retention time of sewage in the WWTP, conventional activated sludge methods of treatment can remove between 85 and 99% of acetaminophen from wastewater (Verlicchi et al. 2012). However, UV irradiation of wastewater during sewage treatment causes the formation of a toxic photoproduct of acetaminophen, 1-(2-amino-5-hydroxyphenyl)ethanone (Kawabata et al. 2012). The risks to human health of long-term, low-level acetaminophen intake, on the other hand, are relatively low. Acceptable daily intake of acetaminophen is at the upper limits for all PPCPs, at 340 µg/kg/day (Schwab et al. 2005), and it is found at an average of about 10 µg/L in streams across the US (Kolpin et al. 2002). Recently, however, chronic use of acetaminophen is being discouraged due to effects on the liver. Acetaminophen has been shown to lower activity levels in *Rana pipiens* tadpoles, especially when caffeine is also present (Fraker and Smith 2004).

Cimetidine is a histamine H2-receptor antagonist used for the treatment of gastrointestinal disorders. It has shown a removal efficiency through primary and secondary wastewater treatment of about 70% (Buth et al. 2007). Chlorination of wastewater effluent causes the formation of multiple cimetidine by-products with increased toxicity (Buth et al. 2007). Lee et al. (2015) examined possible ecological impacts of cimetidine exposure on two daphnid species and the fish *Danio rerio* and found that at relevant environmental levels cimetidine poses no direct impact on aquatic organisms. It is also known to impair the

growth, reproduction, and survivorship of aquatic invertebrates such as water pennies and amphipods at environmentally relevant levels (0.07 – 70.0 ug/L), although it has not shown a significant impact on primary producers (Hoppe et al. 2012). Long-term effects of low-level exposure are unknown and require further investigation.

Diphenhydramine is a common antihistamine used topically, orally, and as an injection to treat minor and severe allergic reactions. It was one of the five most frequently detected substances in a suite of 105 PPCPs tested for at 23 stream sites in Iowa (Kolpin et al. 2004). In Nebraska, significantly higher concentrations of diphenhydramine have been detected in samples downstream of WWTPs, suggesting that conventional wastewater treatment is inadequate for the removal of this PPCP (Bartelt-Hunt et al. 2009). At environmentally relevant concentrations (0.8 ug/L), diphenhydramine has been shown to impair the reproduction of *Daphnia magna* (Berninger et al. 2011). Concentrations of diphenhydramine that are otherwise sublethal to another daphnid, *Ceriodaphnia dubia*, when combined with low levels of the antidepressant sertraline can cause 100% mortality (Goolsby et al. 2013). This highlights the potential of diphenhydramine to become especially dangerous synergistically in mixtures with other PPCPs.

Prescription Medications

Amphetamine is a potent central nervous system stimulant available by prescription to treat activity deficit hyperactivity disorder and narcolepsy, and for weight loss. Amphetamines are also used by humans in some non-prescription drugs. WWTPs with trickling filter beds remove about 95% amphetamine from wastewater (Kasprzyk-Hordern et al. 2009). Also, amphetamine can be completely removed from raw water in DWTPs through the pre-chlorination, flocculation, and sand filtration processes (Huerta-Fontela et al. 2008). However, amphetamine is nonetheless often detected in surface waters (la Farré et al. 2008). At the upper limit of concentrations found in the environment, amphetamine can have negative oxidative and genetic effects on the zebra mussel *Dreissena polymorpha* (Parolini et al. 2016). The ecological and human health impacts of environmental amphetamine have otherwise not been deeply explored (Rosi-Marshall et al. 2015).

Carbamazepine is an anticonvulsant used to treat seizures, nerve pain, and bipolar disorder. It is consumed by a relatively small population compared to many of the other PPCPs listed here, but it has a very low degradation rate (4-8%), meaning that this PPCP does not require a constant source for it to be present in the environment (Corcoran et al. 2010). Its persistence has led to its use as an anthropogenic marker in the aquatic environment (Clara et al. 2004; Fenz et al. 2005). Carbamazepine also has deleterious effects on the feeding behavior and swimming speed of fish (Nassef et al. 2010), and it has been shown to alter enzymatic activity in the fish *Cyprinus carpio* (Malarvizhi et al. 2012).

Morphine is a narcotic used to treat moderate to severe pain. It has a removal rate of over 80% through activated sludge wastewater treatment (Wick et al. 2009). Morphine has been detected downstream of WWTPs at 0.5 - 32.3 ng/L (Baker and Kasprzyk-Hordern 2013), and its presence in aquatic systems is of high concern (Huber et al. 2018). The fungal enzyme laccase is extremely effective at removing aquatic morphine, and it may be used in the future as an additional measure in wastewater treatment, especially from hospitals (Huber et al. 2018). Ecotoxicological impacts include the impairment of feeding and immune function in the mussel species *Elliptio complanata* (Gagné et al. 2006).

Phenazone is a nonsteroidal anti-inflammatory drug and an analgesic commonly used to relieve pain and fevers. In Germany it has been detected at the ug/L level in public water supply and in wells (Nikolaou et al. 2007). The removal efficiency of phenazone in conventional WWTPs is relatively low, around 33%, and it also shows high environmental persistence (Pieper et al. 2010), two factors allowing phenazone to be detected in groundwater, surface water, and drinking water (Zühlke et al. 2004). Phenazone has been found in the tissues of algae, daphnids, and fish, but the specific toxicological impacts to these organisms remain unknown (Sanderson et al. 2003).

Sulfamethoxazole is an antibiotic used to treat bacterial infections in both humans and animals. It is broken down significantly by photodegradation, which may become especially pronounced in wastewater compared to natural water (Andreozzi et al. 2003; Corcoran et al. 2010; Ryan et al. 2011). The ecological impacts of sulfamethoxazole include fostering antibiotic-resistant bacteria (Kim and Aga 2008). It may also cause immobilization of

Daphnia magna, and a toxic photoproduct of sulfamethoxazole has additionally been shown experimentally to increase this effect (Jung et al. 2008; Trovó et al. 2009).

Trimethoprim is an antibiotic used to treat bacterial infections in humans, and it is sometimes combined with sulfamethoxazole in a drug called Sulfatrim. It may foster antibiotic-resistant bacteria in the environment, which is a serious threat to public health (Kim and Aga 2008). This PPCP is broken down significantly by photolysis, especially in wastewater compared to in natural water (Ryan et al. 2011). It can have adverse, acute impacts on both aquatic invertebrate and fish species, although chronic effects are still unknown (Choi et al. 2008).

Recreational Drugs

MDMA (3,4-Methylenedioxymethamphetamine), commonly known as ecstasy, is an addictive psychoactive drug. It has been found in surface waters in Spain, with higher levels occurring on weekends and during winter months (Huerta-Fontela et al. 2008). MDMA showed a low removal efficiency (28%) in a DWTP through an ozonation process, but a granulated activated carbon filter was able to raise this to an 88% removal rate (Huerta-Fontela et al. 2008). MDMA has been detected at the ng/L level in WWTP effluent in Spain, Italy, and the US, and it typically has a removal rate from wastewater of around 50% (Zuccato and Castiglioni 2009). To date there have been no studies regarding the ecological and human health impacts of MDMA at environmentally relevant levels.

MDA (3,4-Methylenedioxyamphetamine) is a psychostimulant and psychedelic drug not commonly used, but it is a metabolite of all amphetamine drugs. It makes up between 8 and 9% of the metabolite products of MDMA (de la Torre et al. 2004; Metcalfe et al. 2010). MDA has been detected in surface waters in Spain, with significant increases in concentrations during weekends and summer months (Huerta-Fontela et al. 2008). In a study on untreated wastewater in Canada, MDA was most often at levels below the detectable limit (Metcalfe et al. 2010). However, it is effectively removed by conventional DWTP processes (Huerta-Fontela et al. 2008). To date there have been no studies regarding the ecological and human health impacts of MDA in the environment.

Methamphetamine is a strong and highly addictive central nervous system stimulant used commonly as a recreational drug and rarely as a treatment for attention deficit hyperactivity disorder and obesity. WWTPs typically remove between 60 and 98% of methamphetamine from wastewater (Zuccato and Castiglioni 2009). However, in a study on Nebraska streams impacted by WWTPs, methamphetamine was detected in every sample but one, and typically at higher levels downstream of WWTPs, in the ng/L range (Bartelt-Hunt et al. 2009). It has also been detected at the ng/L level in DWTP intake water in Spain, but sand filtration can effectively remove this PPCP from water (Huerta-Fontela et al. 2008). Methamphetamine can enhance the long-term memory of the aquatic snail *Lymnaea stagnalis*, demonstrating the concern that should be placed on environmental methamphetamine as well as its potential to cause similar harm to human health (Kennedy et al. 2010).

Agricultural or Veterinary Compounds

Sulfamethazine, or sulfadimidine, is an antibacterial agent used only for livestock in the US (Pérez et al. 2006). Conventional secondary wastewater treatment in WWTPs effectively removes between 80 and 100% of sulfamethazine from wastewater (Yang et al. 2005; Karthikeyan and Meyer 2006; Gao et al. 2012). Kim et al. (2007) did not detect acute responses to sulfamethazine in the marine bacterium *Vibrio fischeri Daphnia magna*, or in the fish species *Oryzias latipes*. Longer term effects of exposure to sulfamethazine include the inhibition of the growth of duckweed (*Lemna gibba*), the mobility and survival of *Daphnia magna*, and the growth and survival of invertebrate *Moina macrocopa* and fish *Oryzias latipes* (Ji et al. 2012).

Sulfachloropyridazine is an antibacterial agent used primarily on livestock. It may be present in manure from treated livestock, which may be applied to land. Conventional WWTPs are able to remove over 93% of sulfachloropyridazine from wastewater (Michael et al. 2013). DWTPs are likewise relatively efficient in the removal of this PPCP, at an average of 90.3% removal (Michael et al. 2013). There are no detectable acute responses to sulfachloropyridazine in *Vibrio fischeri*, *Daphnia magna*, or *Oryzias latipes* (Kim et al. 2007). However, there is evidence that it can cause antibacterial resistance in soil bacteria,

posing large potential threats to public health (Schmitt et al. 2004). Further investigation is needed as to other long-term ecotoxicological impacts of sulfachloropyridazine.

Thiabendazole is a fungicide that has been used to treat produce, Dutch elm disease, and to counter parasites such as roundworms in humans and livestock. It has been detected in agricultural-food production effluent, in soil sediments, and in some aquatic organisms (Castillo et al. 1997). Thiabendazole is one of the most commonly detected pesticides in agricultural-food wastewater, and it is not effectively removed by conventional activated sludge processes or by an advanced membrane bioreactor technique (Sánchez Pérez et al. 2014). The only known agricultural-food wastewater treatment process known to effectively remove thiabendazole is through advanced oxidation (Sánchez Pérez et al. 2014).

Study Area

This study was conducted on, Messalonskee Lake the final and easternmost lake in the chain of the Belgrade Lakes, located in Kennebec County, ME, in the towns of Sidney, Oakland, and Belgrade (Figure 4). It has an area of 1493 hectares, a mean depth of 10 meters, and a maximum depth of 34 meters (Maine Volunteer Lake Monitoring Program 2018). The southern end of the lake is shallower than the central and northern sections of the lake. The water enters the lake from the Belgrade Stream at the southern end and drains at the northern end, and the residence time of the lake is 0.63 years (Colby Environmental Assessment Team 1997). The lake is mesotrophic, and it suffers from outbreaks of the invasive aquatic plant Eurasian watermilfoil (Friends of Messalonskee 2017).

There are three boat launches on Messalonskee Lake, one public beach, and a railroad runs along the western side of the lake. Friends of Messalonskee personnel inspected 1483 boats at the Oakland boat launch and 1392 boats at the Sidney boat launch during the summer of 2017 (pers. comm. Rachel Whitney). The northern end of the lake is more developed than the southern end, which is primarily undeveloped wetland.

The Belgrade Lakes region has throughout the past century undergone increased levels of residential and commercial development, most notably as a destination for seasonal residents and tourists drawn by natural surroundings and outdoor recreation. This

development has certainly increased the number of septic systems around Messalonskee Lake and, thus the amount of effluent discharged in the soils within the Messalonskee watershed (Burgess and Nelson 2009). It is unknown how much septic leakage goes into the lake.

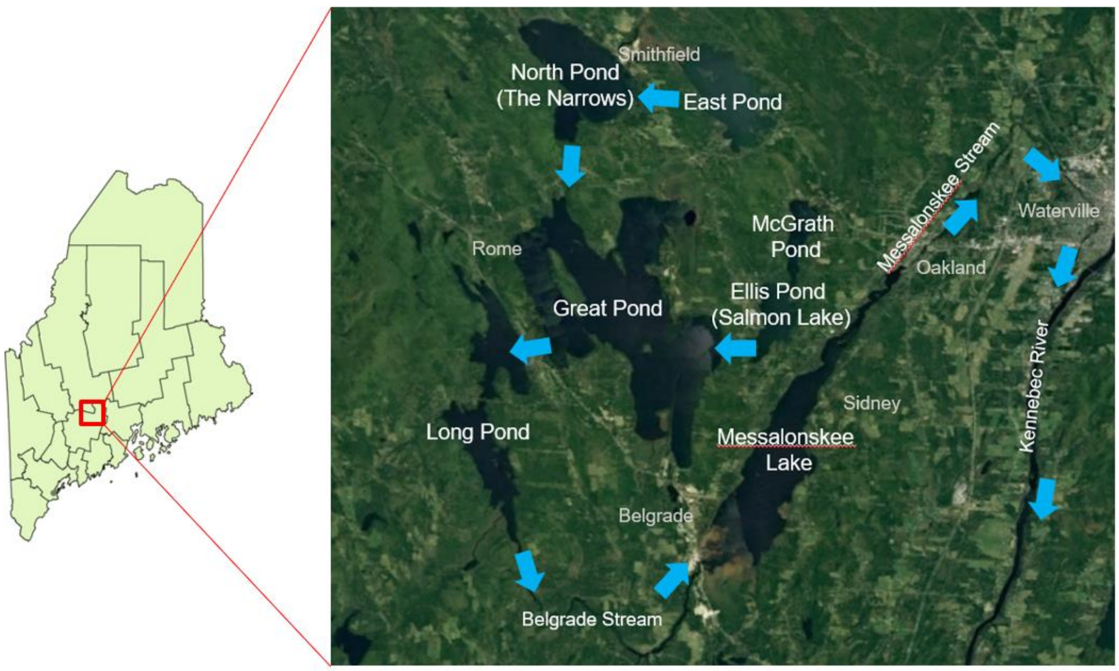

Figure 4. A map of the Belgrade Lakes region, located in Kennebec County and Somerset County, ME. White labels indicate lake names. Tan labels indicate streams and town names. Blue arrows indicate flow direction.

METHODS

Messalonskee Lake Water Sampling and Processing

There are 13 locations on the lake from which we collected samples each week (Figure 5), and we collected a duplicate sample once from each site, for a total of 65 lake samples. Exact locations of all sampling sites and substrate descriptions are provided in Appendix 1. We identified sample locations based on their spatial distribution, human use level, and accessibility. All lake sites were binned into four categories based on human use: open water, private nearshore, public nearshore low use, and public nearshore high use (Table 2). We collected water samples four times between June 26 and August 7, 2017, once every two weeks. We collected samples on Mondays in an attempt to capture potentially higher values of contaminants following higher levels of activity over the weekend.

One of the sites is on Belgrade Stream, the inlet stream to Messalonskee Lake close to where it enters the lake. Sampling at this site allows detection of concentrations of PPCPs in the water coming into the lake. Another site is before the outlet dam and may indicate PPCPs leaving the lake.

We collected samples from the 10 near-shore sites where the water depth was about 1 m, ranging from 4.3 to 11.2 m from shore. We collected open water samples from the bow of a motorized boat, always facing upwind to avoid contamination from the boat. Two of the open water sites were at existing data collection buoys, facilitating relocation each time we sampled. We relocated the third open water site using a Garmin GPS 76. At each site on each sampling date, we measured dissolved oxygen and temperature using an Onset® HOBO® logger. General weather conditions were also recorded.

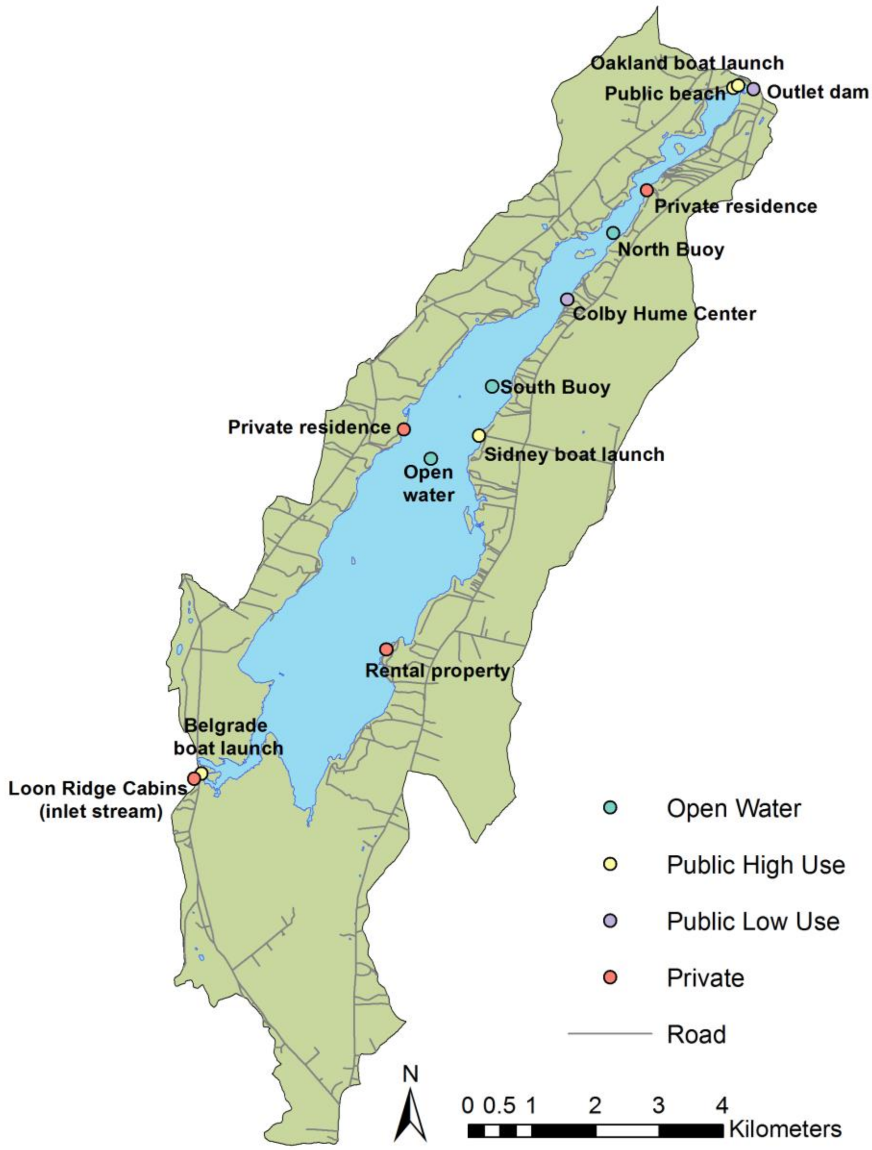

Figure 5. A map of the 13 sampling sites around Messalonskee Lake used in this study, indicating their usage in four colors. The green area represents the Messalonskee Watershed.

Table 2. Descriptions of the 13 sampling sites on Messalonskee Lake (ML), the two sites on Kennebec River (KR), and the one site on Messalonskee Stream (MS).

Site ID	Site description	Level of usage	Dist. from shore (m)	Substrate
ML1	Outlet dam	Public Low	5.6	Gravel, concrete, detritus
ML2	Oakland boat launch	Public High	7.3	Concrete ramp, rip-rap
ML3	Oakland beach	Public High	11.2	Gravel, leaf litter
ML4	North Buoy (MESSDEP 2)	Open Water	--	--
ML5	Middle of South Buoy (MESSDEP 1)	Open Water	--	--
ML6	Middle of lake	Open Water	--	--
ML7	Residence (East)	Private	5.8	Pebbles, muck, rocks
ML8	Hume Center	PublicLow	14.7	Sand, rocks, boulders
ML9	Sidney boat launch	PublicHigh	10.8	Concrete ramp, rip-rap
ML10	Rental property	Private	6.8	Boulders, rocks
ML11	Belgrade boat launch	Public High	5.2	Concrete ramp, muck, milfoil
ML12	Inlet stream	Private	4.3	Rocks, milfoil
ML13	Residence (East)	Private	5.9	Sand, rocks
KR1	River, downstream of WWTP	--	--	Rocky, fast current
KR2	River, upstream of WWTP	--	--	Concrete/rock
MS	North St. boat launch	--	--	Gravel, some large rocks

We also collected duplicate water samples from lotic sites downstream of Messalonskee Lake on August 9, 2017. One site was on Messalonskee Stream at the North Street boat launch in Waterville. The other two sites were on the Kennebec River, one upstream of the Kennebec Sanitary Treatment plant (at the public boat launch in Waterville) and one downstream (Figure 6). The two boat launch sites were tested to compare to the lake sites, and the downstream river site was intended to be a possible "positive" control based on results from other studies of surface water near WWTPs.

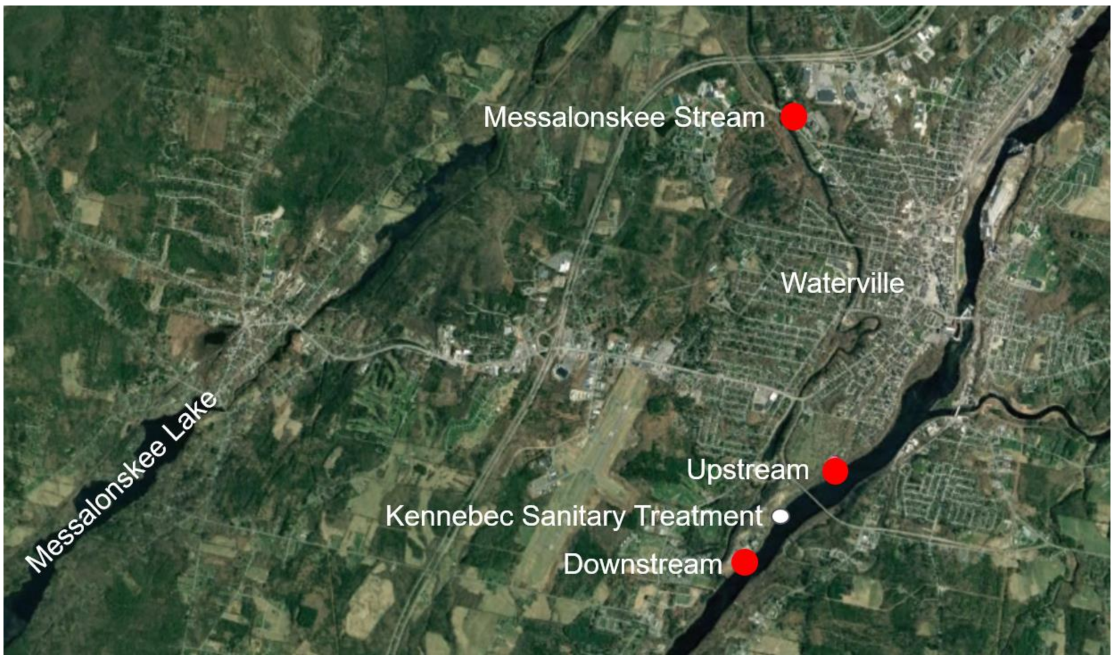

Figure 6. A map of the three non-lake sites, sampled on 8/9/2017. Sampling sites are labelled with red circles, and the location of the Kennebec Sanitary Treatment plant is indicated with a white circle. The northern end of Messalonskee Lake is indicated in the southwest corner of the map, which flows out into the Messalonskee Stream, flowing northeast before curving southeast heading into Waterville. Messalonskee Stream flows enters the Kennebec River downstream of the Kennebec Sanitary Treatment Plant.

We did not consume/apply any of the substances being tested prior to sampling or processing. We wore nitrile gloves during sampling and processing to prevent possible contamination. We collected all samples for chemical detection in 1 L, amber glass bottles rinsed with acetone and Milli-Q. We rinsed bottles three times with lake, stream, or river water before filling them from about 15 cm below the surface by inverting the bottle while lowering it to sampling depth. Samples were immediately stored on ice and were refrigerated within six hours of collection.

We performed solid phase extraction (SPE) to prepare the samples for liquid chromatography-mass spectrometry (LC-MS) no more than three days after collection (Batt and Aga 2005; Batt et al. 2006; Roberts and Thomas 2006). Using vacuum filtration, we ran the samples through a 25 mm GMF (glass microfiber, 1µm nominal pore size) filter into a 500-mg Oasis HLB cartridge (Waters, Milford, MA). Prior to running each sample, we conditioned the system with 5-10 mL of methanol followed by 5-10 mL of Milli-Q water. We processed 100-500 mL of each sample at an average flow rate of 9 mL/min.

Afterwards, we removed the cartridges and froze them at -70 °C until all samples were prepared for shipment to the Water Sciences Laboratories at the University of Nebraska (Daniel Snow, Director). Samples were then analyzed for a suite of 18 different PPCPs using liquid chromatography-tandem mass spectrometry followed by positive and negative ion detection. Specific PPCPs tested for were determined by the Water Sciences Lab. A detailed description of these analytical methods is provided by Bartelt-Hunt et al. (2009).

Orphenadrine was used to measure potential interference of dissolved organic carbon (DOC) concentrations with PPCP concentrations. DOC was not measured during our study, and due to a gap of about three months between sampling and receiving the analyzed data from the Water Sciences Lab, it was not possible to deeply explore the variability in orphenadrine used. The range of percent orphenadrine ranged between 33.4% and 217.3%, and the mean was 98.5%.

The 18 compounds for which each sample was tested, all commonly found in surface waters, include three stimulants (caffeine, 1,7-dimethylxanthine, and cotinine), three over-the-counter medications (acetaminophen, diphenhydramine, and cimetidine), three

agricultural or veterinary chemicals (sulfamethazine, sulfachloropyridazine, thiabendazole), six prescription medications (sulfamethoxazole, trimethoprim, carbamazepine, amphetamine, morphine, and phenazone), three recreational drugs and (MDMA, MDA and methamphetamine; Table 1).

On July 24 and August 7 we also collected samples for total phosphorus and total nitrogen analyses. We collected these samples in 60 mL acid-washed plastic bottles in duplicate from each of the 13 lake sampling sites. We stored the samples frozen until analyzed. For total phosphorus, we followed the standard operating procedure for total and dissolved phosphorous (LaChat Method 10-115-01-1-F for QuikChem FIA+8000). For total nitrogen, we followed the standard operating procedure for particulate-phase total nitrogen by alkaline persulfate oxidation digestion (Lachat Method).

We used ArcGIS to display results by location.

Data Analysis

Since the PPCP concentration data were non-normally distributed, we employed general linear models (GLM) in R (v. 3.4.2) to compare factors such as site, site use, weather, nutrient levels, and sampling date. Depending on the data involved in each test, we used the families “poisson” and “binomial.” We followed GLMs with a post-hoc general linear hypothesis test, which is used to determine groups that are significantly different when making multiple comparisons for GLMs.

Septic System and Drinking Water Survey

We created an online survey using Qualtrics for Messalonskee watershed residents to answer questions about their septic systems and drinking water sources (Appendix 2; Qualtrics, Provo, UT). The survey was approved by the Colby Institutional Review Board for Research with Human Subjects before being released to the public. The survey was distributed to the public by word of mouth, on the Friends of Messalonskee website and Facebook page, on flyers posted in local businesses, and through a hard copy version of the survey provided to the town offices of each of the three towns surrounding Messalonskee

Lake (Oakland, Sidney, and Belgrade). We plotted survey response information on maps of the Messalonskee Lake watershed using ArcGIS.

There were two primary purposes of the survey. The first was to gather information about the condition of septic systems in the Messalonskee Watershed, which may be a source of PPCPs. The second was to collect information on how residents source their drinking water, with attention to the potential public health concerns if residents take their drinking water directly from the lake, which had been suggested to us by residents anecdotally.

RESULTS

Over the four weeks of sample collection on Messalonskee Lake during the summer of 2017, we collected 65 samples that were tested for PPCPs (13 sites x 4 weeks, plus one duplicate for each site gathered either weeks 1, 2, 3, or 4). Lake sampling sites included four public high-use sites (Oakland public beach, Oakland boat launch, Sidney boat launch, Belgrade boat launch), two public low-use sites (outlet dam, Hume center), four private sites (two private residences, a rental property, and the Loon Ridge Cabins), and three open water sites (two MESSDEP buoys and one other open water site; Figure 4).

Environmental conditions, including weather, for each sampling date at each site are in Appendix 3. Dissolved oxygen was not significantly different across the four sampling dates; however, it was significantly different between some of the sampling sites in the lake (Table 3). ML11 had significantly lower dissolved oxygen than every other sampling site on the lake ($p < 0.0001$). ML12 also had significantly lower dissolved oxygen than ML13 ($p = 0.02$).

Temperature did not vary significantly between sampling sites, but it did vary significantly by sampling week (Figure 7). The first week had a significantly lower average temperature than the second ($p < 0.0001$), third ($p < 0.0001$), and fourth weeks ($p = 0.008$).

Five of the 18 PPCPs included in the analysis were detected ≥ 0.005 µg/L detection limit across the 13 sites (Table 4). These include caffeine and its metabolite 1,7-dimethylxanthine, amphetamine, acetaminophen, and sulfachloropyridazine. PPCPs not detected included carbamazepine, cimetidine, cotinine, diphenhydramine, MDA, MDMA, methamphetamine, morphine, phenazone, sulfamethazine, sulfamethoxazole, thiabendazole, and trimethoprim. The complete table of PPCP concentrations detected in each sample can be found in Appendix 4.

Table 3. Dissolved oxygen (DO) and temperature readings averaged over the four sampling dates, by sampling site.

Location ID	Location Name	DO % saturation	DO concentration (mg/L)	temp (°C)
ML1	Outlet dam	99.1 ±2.1	8.2 ±0.2	24.9 ±0.5
ML2	Oakland boat launch	100.4 ±1.8	8.3 ±0.2	25.0 ±0.4
ML3	Oakland public beach	101.0 ±1.8	8.3 ±0.2	25.1 ±0.4
ML4	North buoy	103.2 ±2.0	8.6 ±0.2	24.7 ±0.5
ML5	South buoy	102.4 ±1.8	8.7 ±0.2	23.8 ±0.4
ML6	Open water	101.9 ±1.8	8.6 ±0.2	23.7 ±0.5
ML7	Private residence (East)	102.3 ±2.7	8.5 ±0.3	24.9 ±0.4
ML8	Colby Hume Center	104.2 ±2.0	8.7 ±0.2	24.4 ±0.4
ML9	Sidney boat launch	102.6 ±1.0	8.7 ±0.2	23.9 ±0.5
ML10	Rental property	100.4 ±0.4	8.6 ±0.2	23.4 ±0.4
ML11	Belgrade boat launch	31.1 ±1.2	2.7 ±0.1	23.2 ±0.4
ML12	Loon Ridge Cabins	91.9 ±2.3	7.7 ±0.2	24.3 ±0.4
ML13	Private residence (West)	104.7 ±1.9	8.8 ±0.2	24.2 ±0.5

A sample and its duplicate taken from the Colby Hume Center (ML8) on 6/26/2017 varied in the detected level of amphetamine (0.003 µg/L and 0.024 µg/LBecause one of the two measurements was below the limit of detection, this sample and duplicate were removed from the analysis. All other duplicates matched up, although most were below LOD or zero.

The sample collected from the Kennebec River downstream of Kennebec Sanitary Treatment was intended to be a positive control for PPCP detection. Only two chemicals were detected at this site: acetaminophen and caffeine (Table 5). Water downstream of the WWTP had higher levels of PPCPs than those observed upstream. Caffeine was detected at over double the upstream concentration, and acetaminophen was detected an order of magnitude higher. These detection levels were within the range of concentrations observed in Messalonskee Lake for each of the PPCPs (Table 4). Caffeine at 0.035 µg/L was also detected in the sample from Messalonskee Stream at the North St. boat launch in Waterville. This level is higher than any of the caffeine detections from Messalonskee Lake and the Kennebec River.

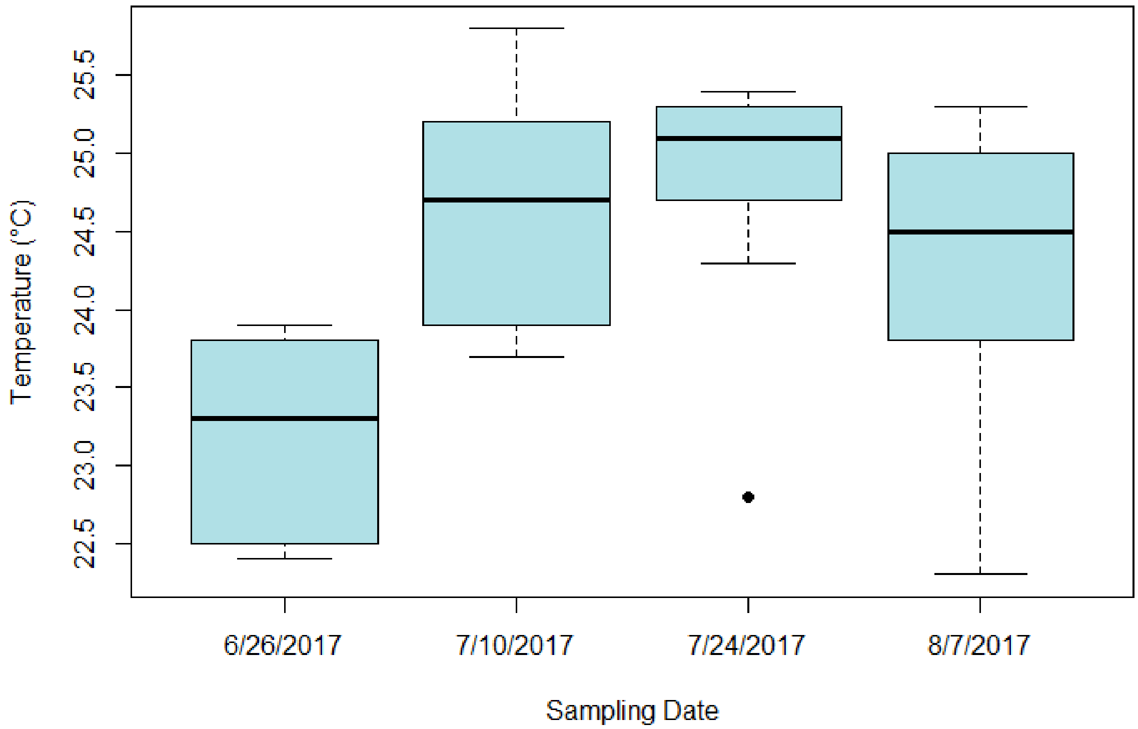

Figure 7. Side-by-side boxplots representing surface (0.5 m) water temperature at each lake site by sampling week. The first week had a significantly lower average temperature than the second ($p < 0.0001$), third ($p < 0.0001$), and fourth weeks ($p = 0.008$).

Temporal Dynamics

Over the six weeks we collected samples, the total count of PPCPs detected ≥ LOD across all sites decreased from 12 the first week to only 1 the last week (Figure 8). Amphetamine individually follows this pattern across the four weeks, although there is variation in this pattern among the other chemicals (caffeine, 1,7-dimethylxanthine, sulfachloropyridazine and acetaminophen). PPCPs were detected at eight sites the first week of sampling, five the second week, three the third week, and only one the final week of

sampling (Figure 9). The fourth week of sampling had a significantly lower count of PPCPs per site than the first week of sampling (Figure 10; $p = 0.091$).

The number of PPCPs detected at each site was not significantly related to the number of days it had rained up to three days before sampling. We considered weather weather was a significant predictor of PPCP detection, looking at weather records the three days before sample collection (data not shown). We did not find a significant relationship between weather and PPCP detection.

Table 4. Average and maximum concentrations for the five detected compounds in Messalonskee Lake. Only includes detections ≥ the limit of detection (≥ 0.005 µg/L). All concentrations are in µg/L.

	Caffeine	1,7-Dimethylxanthine	Amphet-amine	Acetami-nophen	Sulfachloro-pyridazine
# ≥ LOD Detections	4	1	11	3	6
Average	0.011	0.021	0.040	0.006	0.006
Std. Error	0.0063	0.0036	0.0630	0.0012	0.0004
Minimum (≥ LOD)	.005	0.018	0.006	0.006	0.005
Maximum	0.020	0.023	0.236	0.008	0.007

Table 5. Concentrations of the two detected compounds at the Kennebec River sampling sites. Only includes detections ≥ the limit of detection (≥ 0.005 µg/L). All concentrations are in µg/L.

	Caffeine	Acetaminophen
Upstream	0.008 ± 0.000	Below LOD
Downstream	0.018 ± 0.001	0.011 ± 0.001

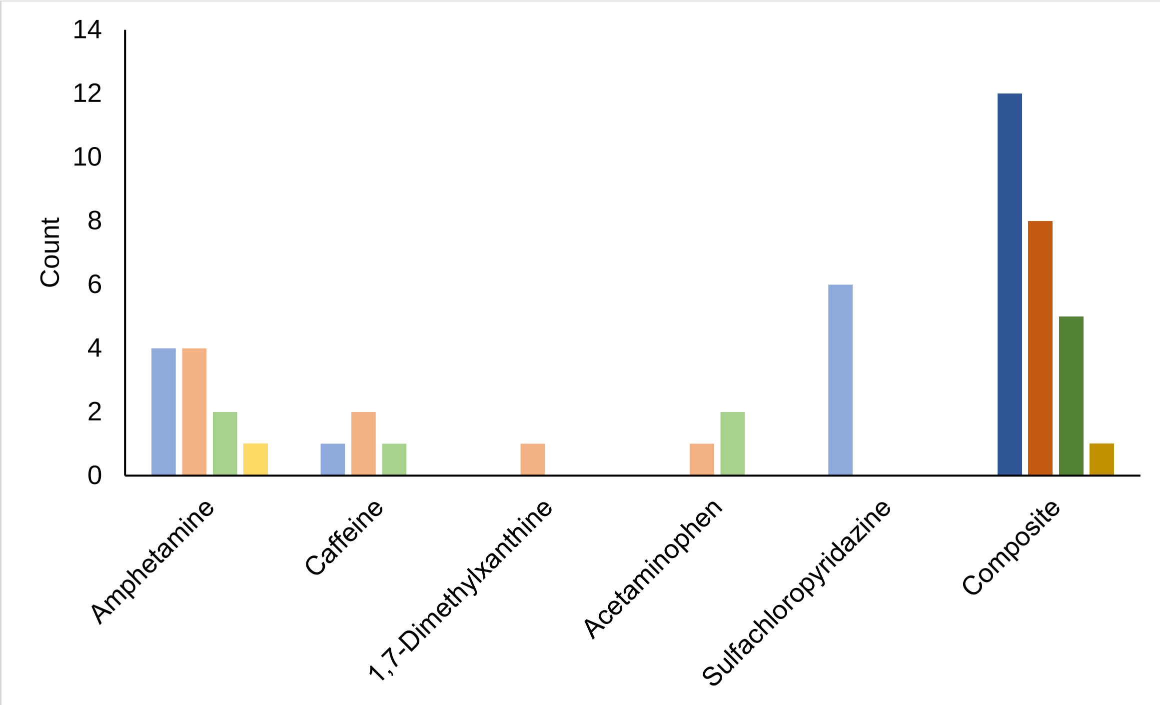

Figure 8. A bar chart showing total number of detections ≥ LOD of each of the 5 detected PPCPs at all sites for each of the four sampling days on Messalonskee Lake, as well as a composite of all positive detections per week. Blue = 6/24/2017; red = 7/10/2017; green = 7/24/2017; yellow = 8/7/2017.

Spatial Dynamics

Refer to Figure 4 in Methods for a map of sample sites and human use categorizations. PPCPs were detected at least once at every site except for the two private residences and the Colby Hume Center (Figure 11), although one of the Colby Hume Center duplicates did have a positive detection for amphetamine, which was discarded because it did not match the other sample. The number of sites with positive detections decreased throughout the four weeks, and the Oakland public beach (ML3) and the Belgrade boat launch (ML11) having the strongest representation throughout the four weeks (Figure 7).

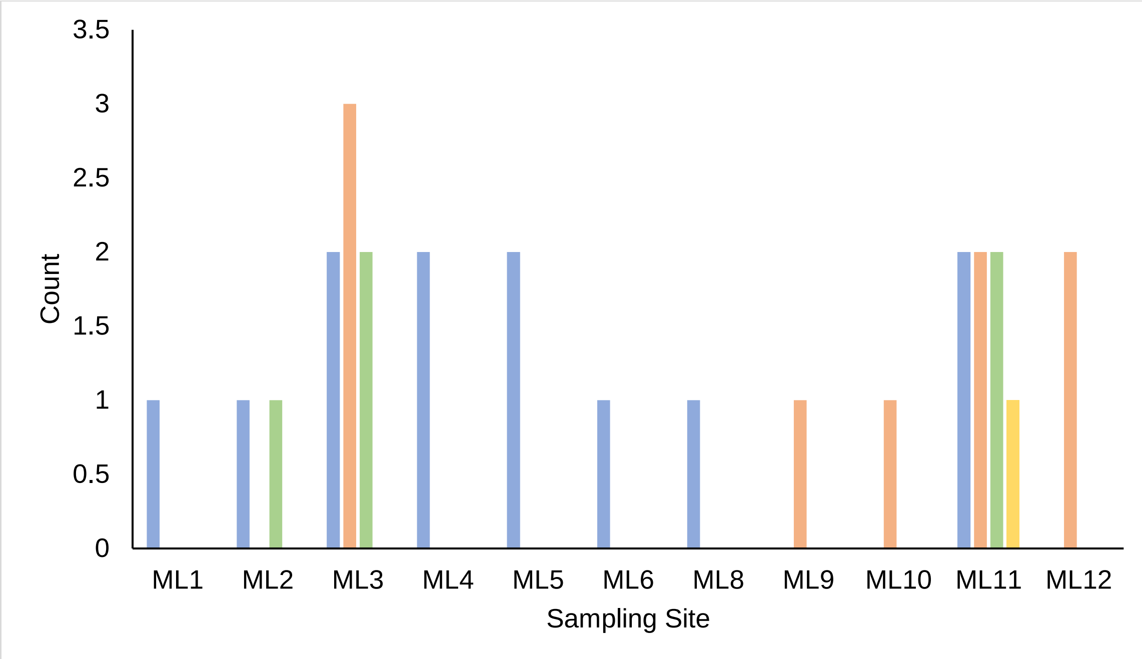

Figure 9. A bar chart showing total number of detected PPCPs (≥ 0.005 μg/L) at each sampling site for each of the four sampling dates at Messalonskee Lake. Blue = 6/24/2017; red = 7/10/2017; green = 7/24/2017; yellow = 8/7/2017. Site names and descriptions are listed in Table 2. ML7 and ML13 (both private residences) and ML8 (the Colby Hume Center) are excluded from the bar chart because PPCPs were never detected at either of these sites.

Breaking up the total count of PPCP detections at each site into detections of each chemical reveals the spatial distribution of specific PPCPs in the lake. Caffeine was detected the most times at the Belgrade boat launch at the south end of the lake and once at the Oakland public beach at the north end of the lake (Figure 12). Its metabolite 1,7-dimethylxanthine was only detected once, at the Oakland public beach (Figure 14). Acetaminophen was only detected at the northern end of the lake, twice at the Oakland public beach and once at the Oakland boat launch (Figure 16a). Sulfachlorpyridazine was only detected on the first sampling date (6/26/2017) at six sites, including the three sites at the northern end of the lake and the three open water sites (Figure 17a). Amphetamine was

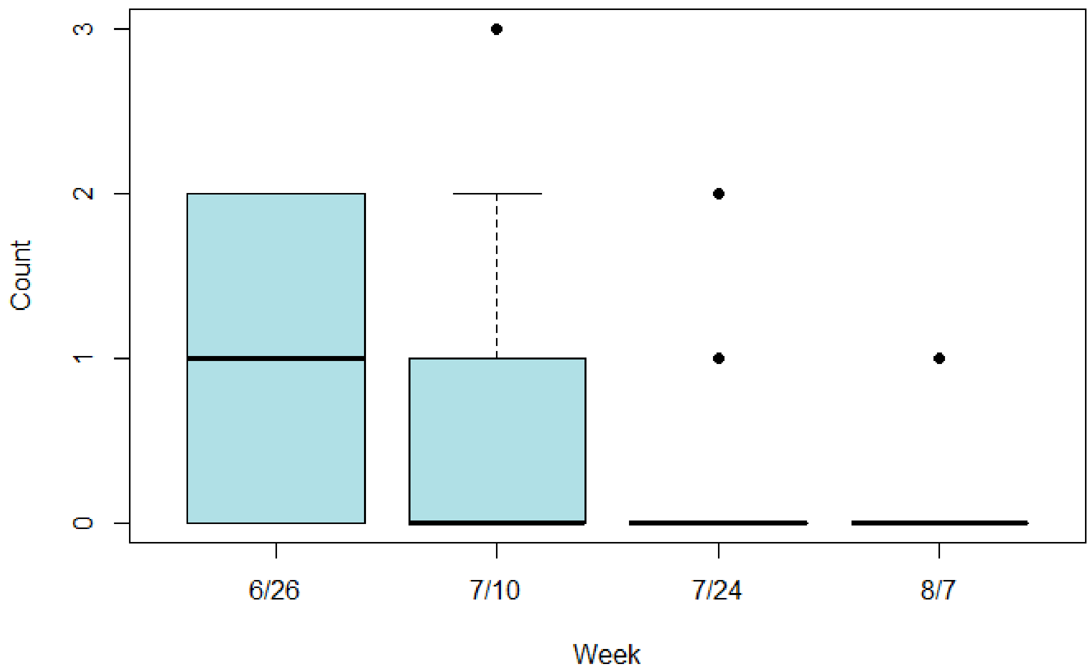

Figure 10. Side-by-side boxplots comparing the average number of PPCPs detected per site over each of the four weeks of sampling. The fourth week of sampling had a marginally significantly lower count of PPCPs per site than the first week of sampling ($p = 0.091$).

detected at many sites throughout the lake, and it was detected the most times at the Belgrade boat launch (Figure 18).

Looking at the maximum concentration of each PPCP at each site, caffeine had its highest detection, at 20 ng/L, at the Oakland public beach although it was detected at a higher frequency at the Belgrade boat launch (Figure 13a). 1,7-dimethylxanthine was detected only once, at the Oakland public beach, and at an even higher maximum concentration than caffeine, at 21 ng/L (Figure 14). Acetaminophen had relatively low maximum concentrations at the two sites at which it was detected, between 5 and 8 ng/L (Figure 16b). Sulfachloropyridazine likewise had low maximum concentrations at all of the sites at which it was detected, just over the limit of detection at 7 ng/L (Figure 17b). The maximum

concentrations of amphetamine were all between 5 and 25 ng/L, with the notable exception of the Belgrade boat launch, where the maximum concentration was a whole order of magnitude higher than any other detection, at 236 ng/L (Figure 19a).

Averaging the concentrations of each PPCP at each sampling site reveals more about the distribution of concentrations across the four sampling dates. The average concentration of caffeine never exceeded 8 ng/L, and it was above the limit of detection at each of the two sites at which it was detected (Figure 13b). The average concentration of 1,7-dimethylxanthine at the one site at which it was detected was 9 ng/L, interestingly higher than any of the average caffeine concentrations (Figure 15). The average concentration of amphetamine is at or just above the limit of detection (5 – 8 ng/L) at the southern buoy, and it is 0.88 ng/L at the Belgrade boat launch, driven up by the sample with 0.236 ng/L amphetamine (Figure 19b). The average concentrations of both acetaminophen and sulfachloropyridazine is below the limit of detection at every sample site and so are not represented in the maps here.

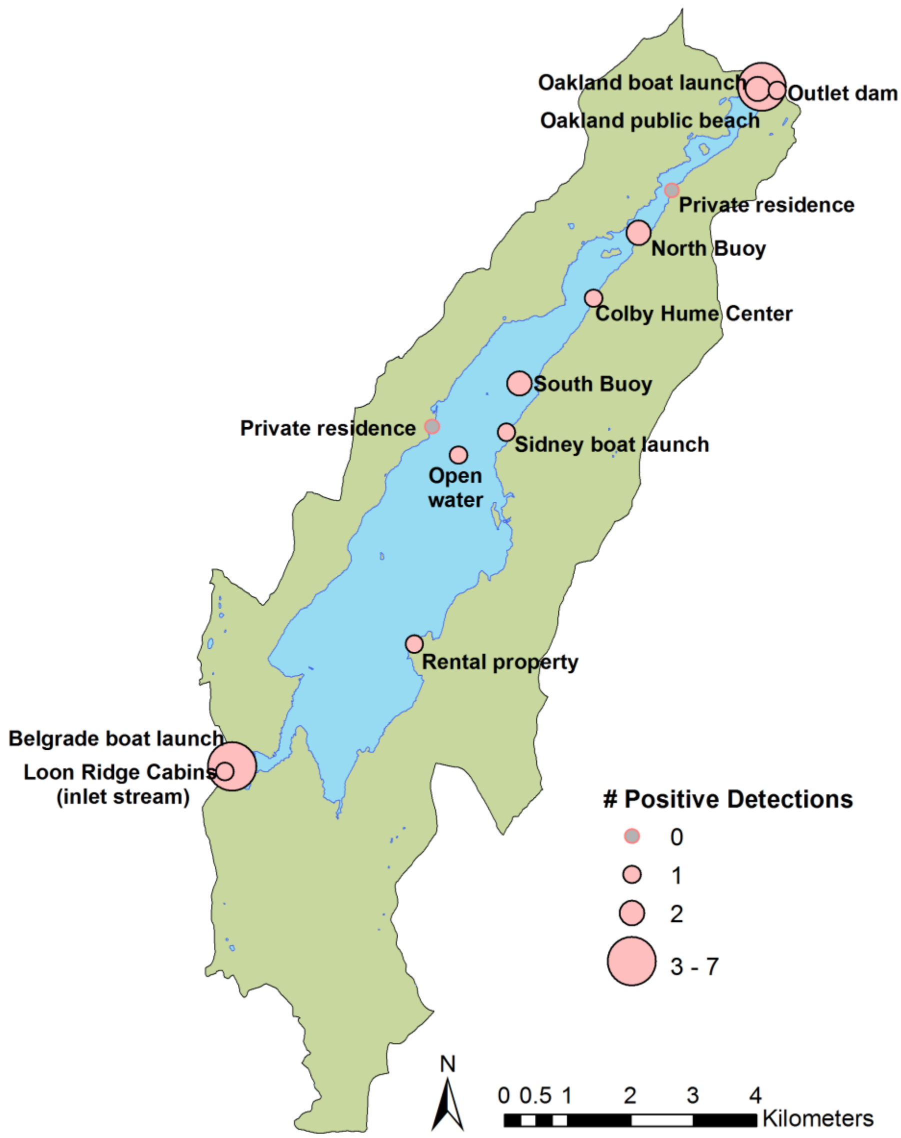

Figure 11. A map representing the total count of PPCP detections (≥ 0.005 μg/L) by sampling site for all four weeks of sampling.

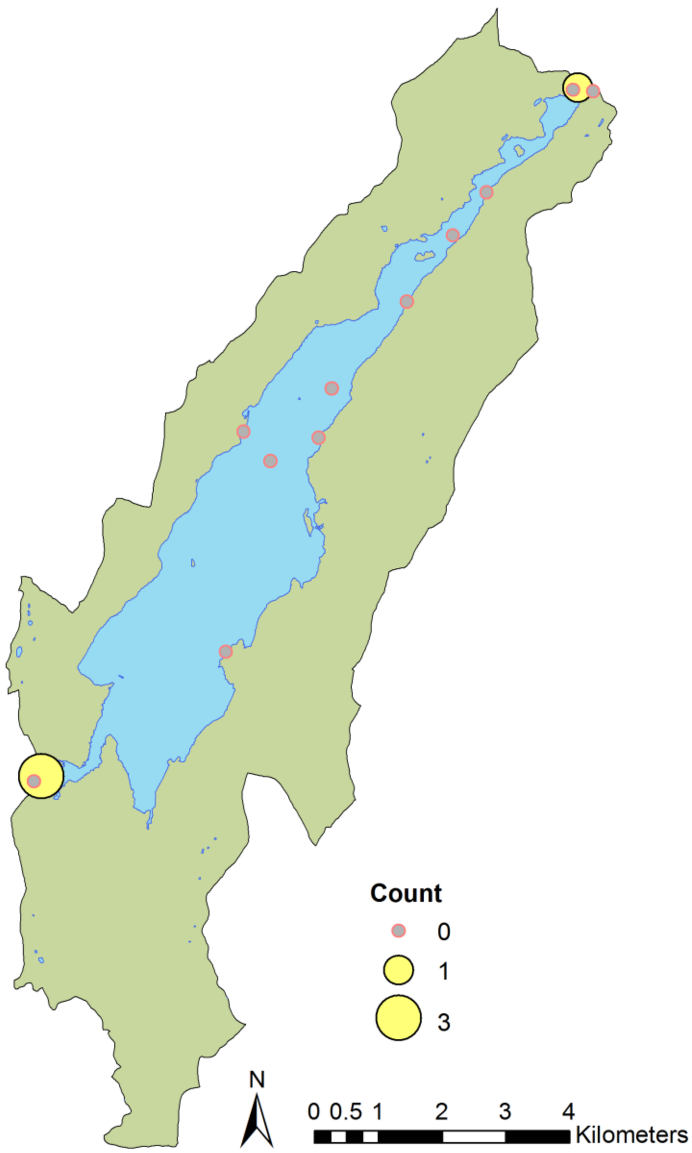

Figure 12. A map of the total number of caffeine detections at each site on the lake (n = 4).

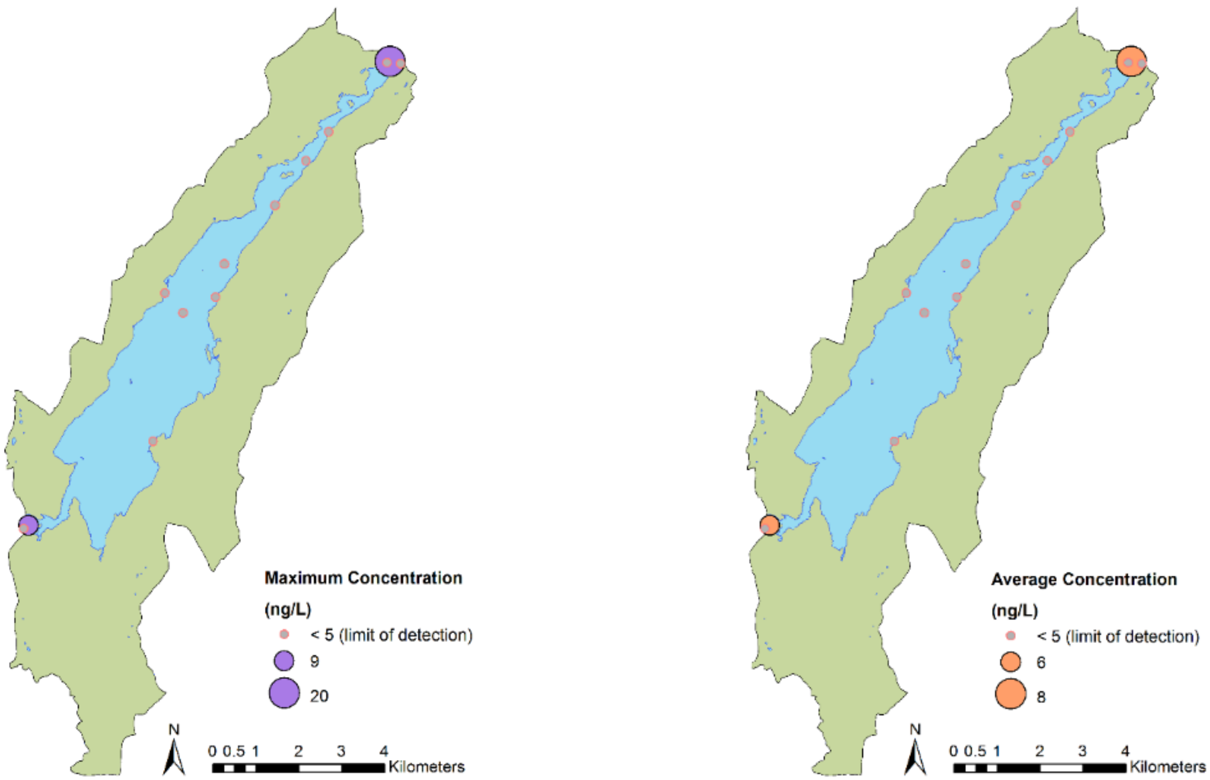

Figure 13. (A) A map of the maximum concentration of caffeine observed at each sampling site. (B) A map of the average concentration (ng/L) of caffeine observed at each sampling site (n = 4).

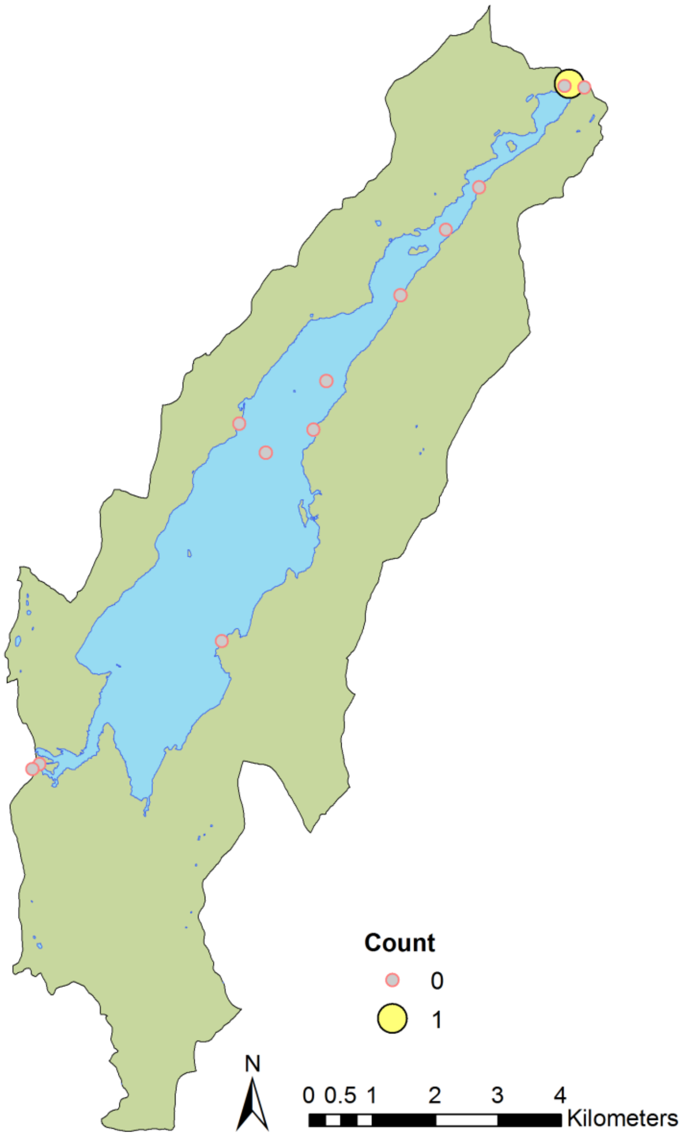

Figure 14. A map of the total number of 1,7-dimethylxanthine detections at each site on the lake (n = 4).

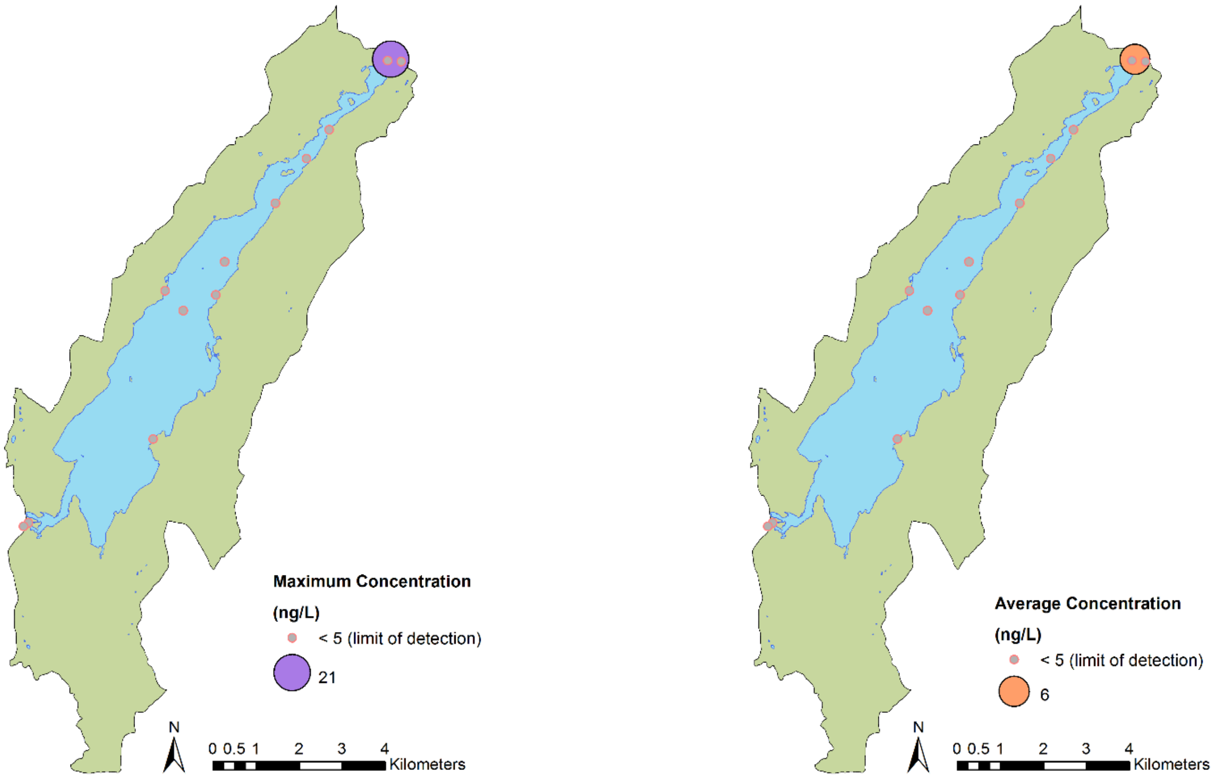

Figure 15. (A) A map of the maximum concentration of 1,7-dimethylxanthine observed at each sampling site. (B) A map of the average concentration (ng/L) of 1,7-dimethylxanthine observed at each sampling site (n = 4).

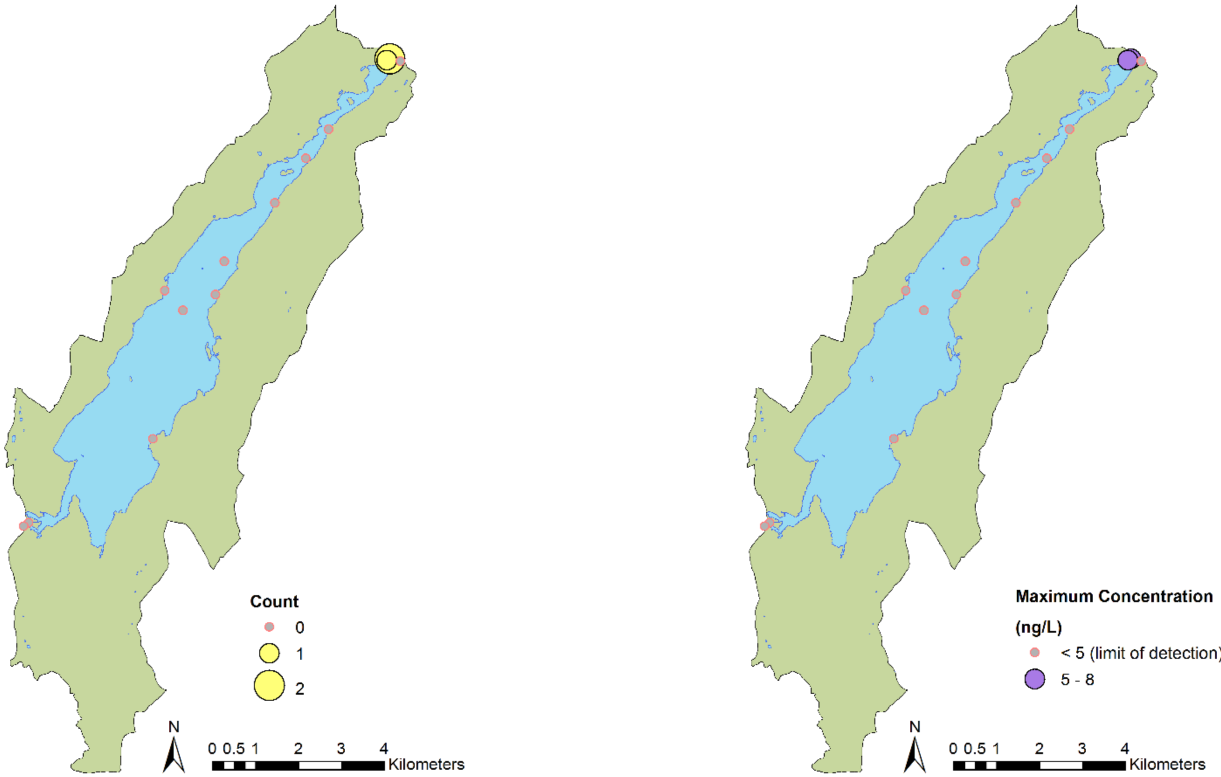

Figure 16. (A) A map of the total number of acetaminophen detections at each site on the lake (n = 4). (B) A map of the maximum concentration of acetaminophen observed at each sampling site.

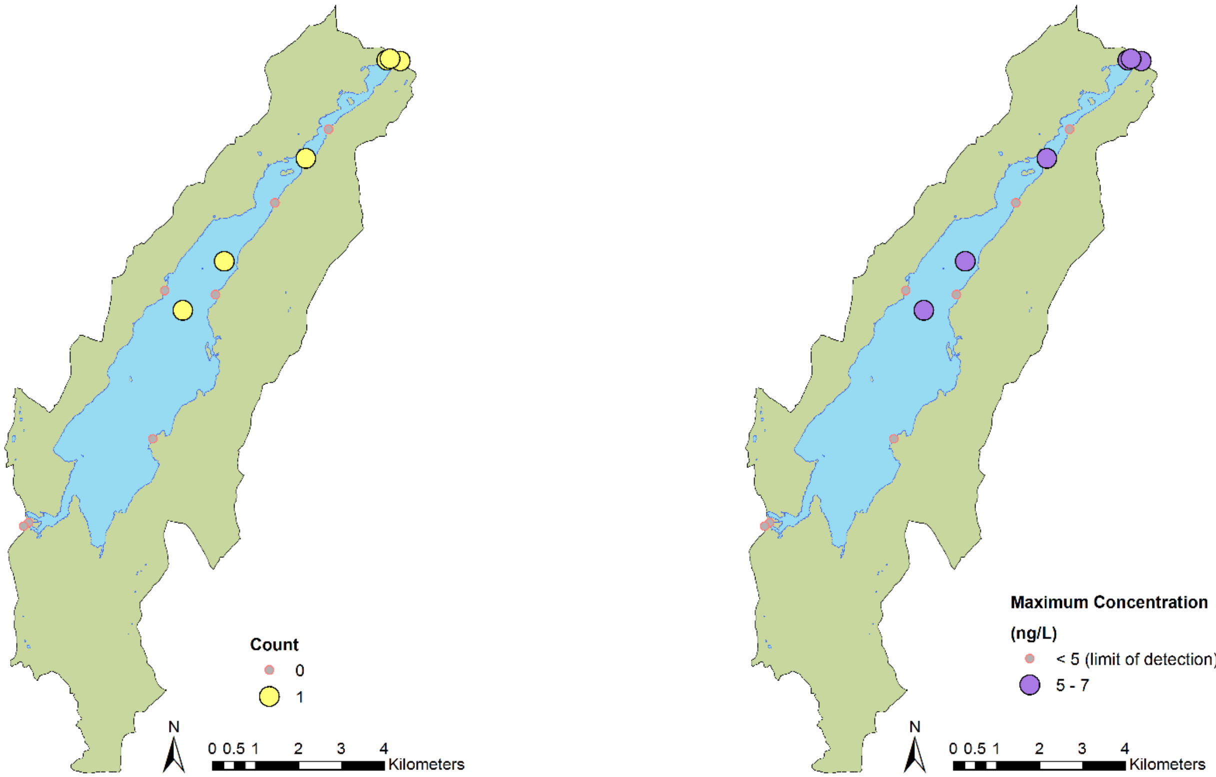

Figure 17. (A) A map of the total number of sulfachloropyridazine detections at each site on the lake (n = 4). (B) A map of the maximum concentration of sulfachloropyridazine observed at each sampling site.

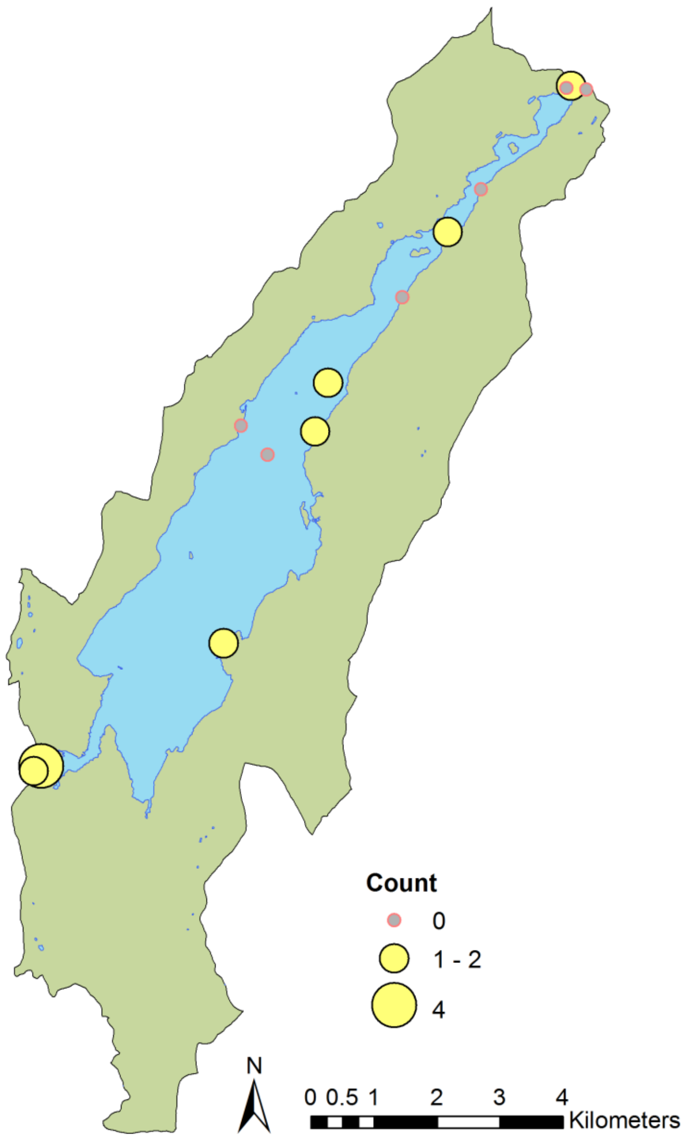

Figure 28. A map of the total number of amphetamine detections at each site on the lake (n = 4).

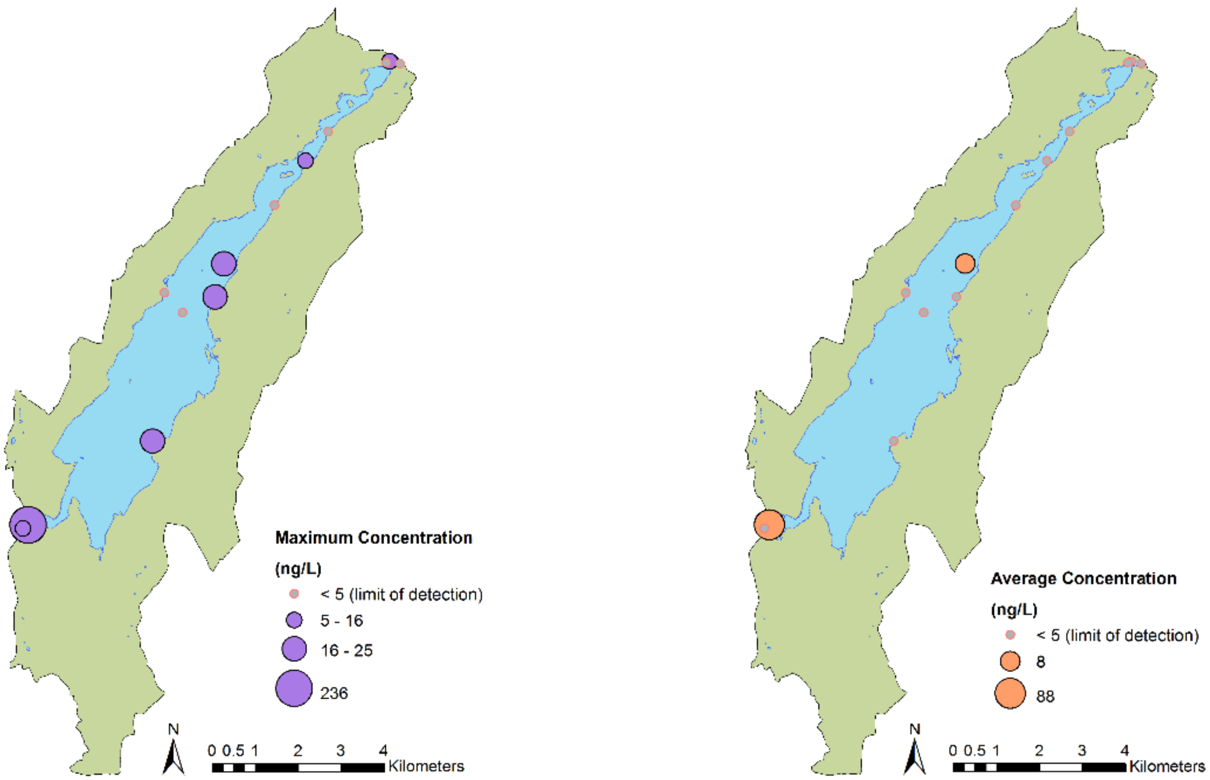

Figure 19. (A) A map of the maximum concentration of amphetamine observed at each sampling site. (B) A map of the average concentration (ng/L) of amphetamine observed at each sampling site (n = 4).

When sites are grouped by human use type (Figure 4), there is a clear difference in the number of PPCP detections (Figure 20). While 63% of samples taken at public high use sites had positive PPCP detections, only 13% of samples taken from private sites had positive PPCP detections. Indeed, the number of PPCPs detected at public high use sites was significantly greater than the number of PPCPs detected at private sites ($p = 0.020$; Figure 21).

Because the total number of detections per sample is low (Table 3), it was not possible to perform robust statistics for individual PPCPs and their concentrations with the exception of amphetamine. Amphetamine was detected more than twice as much as any other PPCP ($n = 11$). We used the natural log of ≥ LOD amphetamine concentration (to normalize) and compared it by site type using a linear model; there were no significant differences, although there were few positive detections at private or open water sites. Variability in amphetamine concentrations, however, was much greater at public high-use sites.

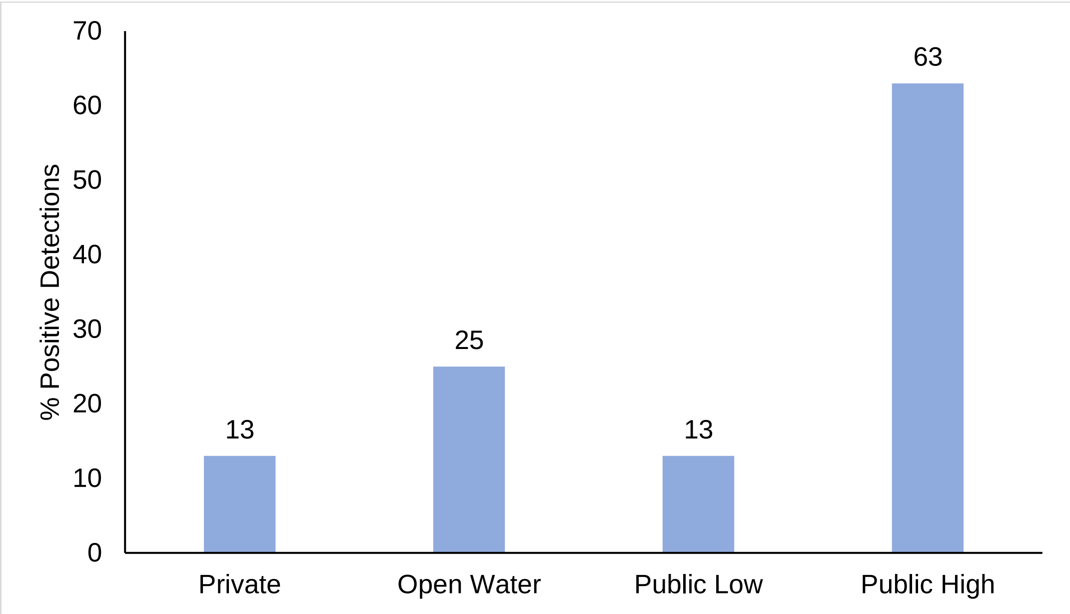

Figure 20. A bar chart comparing the percent of samples that showed positive detections (≥ LOD) of at least one PPCP by human use categories. $n_{private} = 16$; $n_{open\ water} = 12$; $n_{public\ low} = 8$; $n_{public\ high} = 16$.

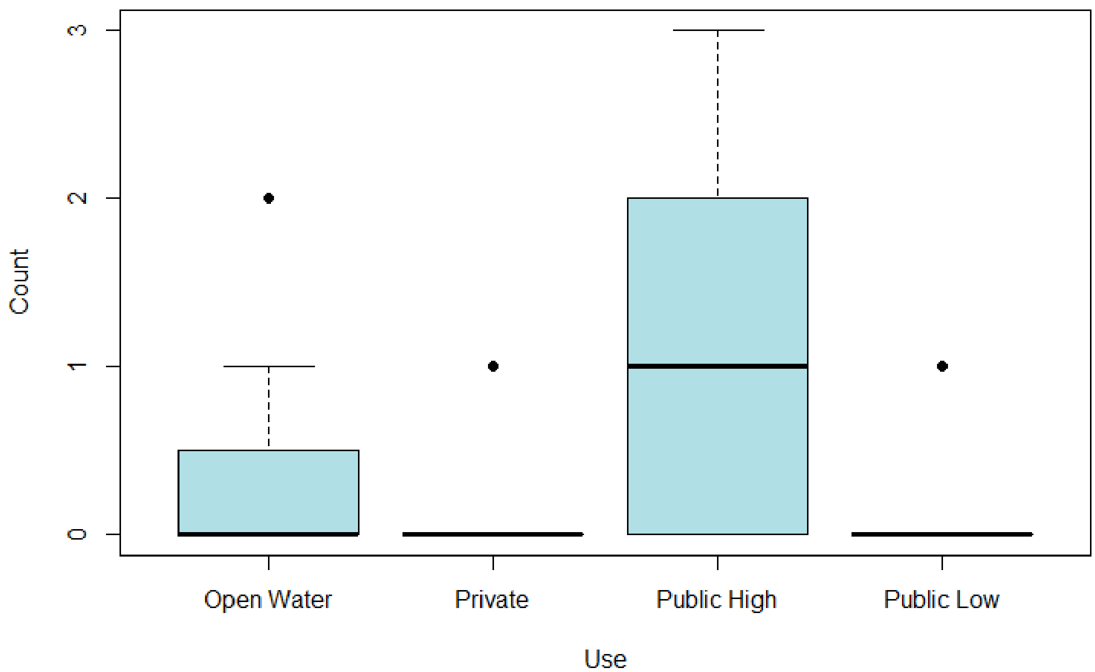

Figure 21. Side-by-side boxplots comparing the total count of PPCPs detected per site across four site use types. Public high use sites had a significantly higher count of PPCPs per site than private use sites (p = 0.020).

Nutrients

We sampled each site for total phosphorus and total nitrogen as a possible measure of the health or level of degradation of each site. Total nitrogen and total phosphorus data organized by sample site can be found in Appendix 5. Total nitrogen (Figure 22) and total phosphorus (Figure 23) varied little among the sites, except the average concentration of total nitrogen observed at the Belgrade boat launch (ML11) was marginally significantly higher than the concentration at one of the private residences (ML10; p = 0.081; Figure 24). Additionally, the average concentration of total phosphorus at the Belgrade boat launch was marginally significantly higher than the concentration at both one of the private residences

(ML10; $p = 0.076$) and the Oakland boat launch (ML2; $p = 0.083$; Figure 22). These relationships are under only a 90% confidence interval.

Total nitrogen and total phosphorus did not correlate significantly to each other. The number of PPCPs detected per sample site did not correlate significantly to total phosphorus measured at that site. However, the number of PPCPs detected per sample site had a significant positive correlation to total nitrogen (Figure 24). Neither total nitrogen nor total phosphorus differed significantly between littoral (near shore) and pelagic (open water) sites across the two dates on which nutrient samples were collected (July 24 and August 7).

Additionally, we gathered Summer 2017 temperature and dissolved oxygen data from the Lake Science Project on the Belgrade Lakes to examine stratification dynamics across our sampling season (pers. comm. B. Fekete). The data, from May 18, 2017 to November 3, 2017, show the lake moving between the end of the spring mixing event to maximum stratification around August 4 (Figure 25). Our sampling dates fall within the period of strong stratification in the lake, when the surface temperatures are the warmest and mixing between the layers is very minimal.

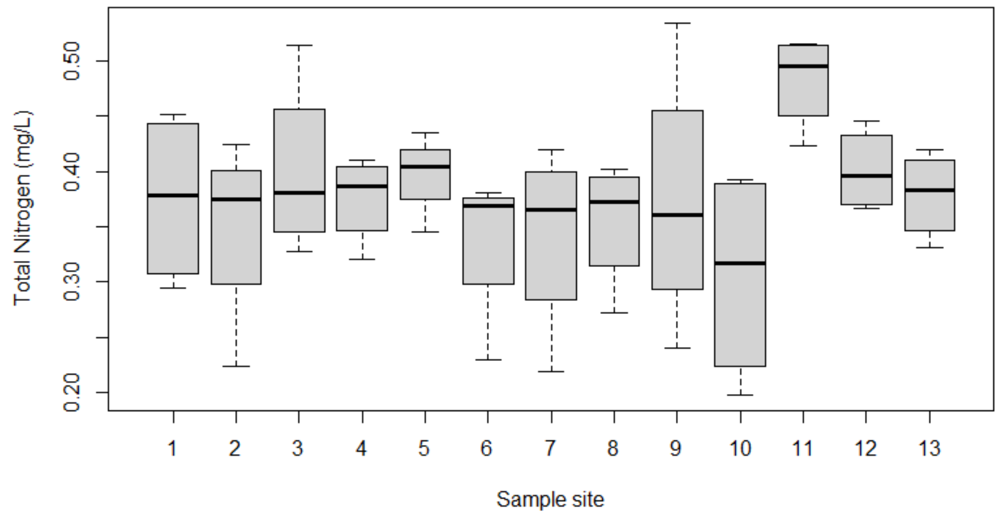

Figure 22. Side-by-side boxplots of total nitrogen (mg/L) by lake sample site (MLx). ML11 (Belgrade boat launch) had marginally significantly higher total nitrogen than ML10 (private residence; under a 90% confidence interval; $p = 0.081$). Differences in total nitrogen between sites were otherwise not significant.

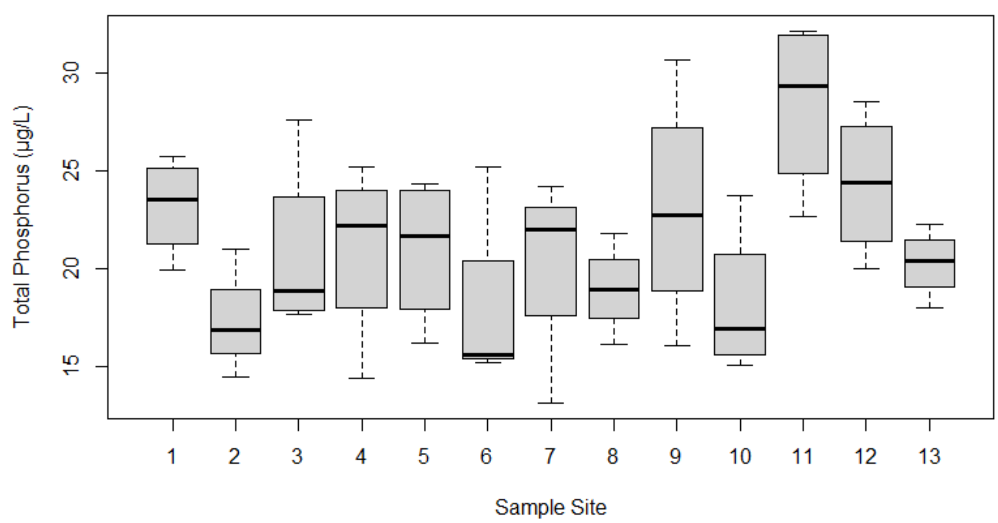

Figure 23. Side-by-side boxplots of total phosphorus (µg/L) by lake sample site (MLx). ML11 (Belgrade boat launch) had marginally significantly higher total phosphorus than both ML10 (private residence; under a 90% confidence interval; $p = 0.076$) and ML2 (Oakland boat launch; under a 90% confidence interval; $p = 0.083$). Differences in total phosphorus between sites were otherwise not significant.

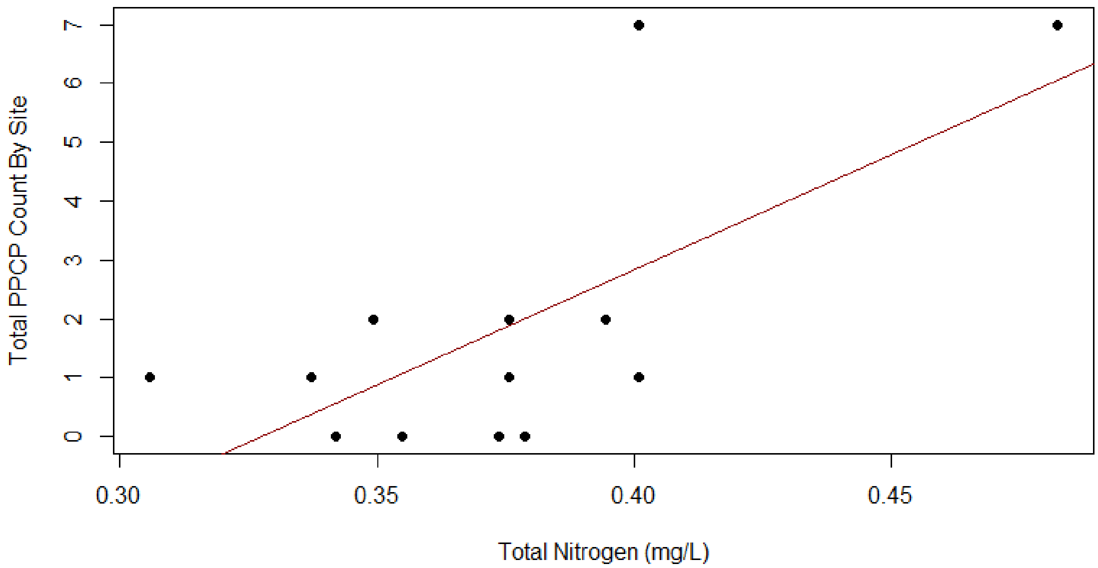

Figure 24. A scatterplot with regression line ($y = 39.2x - 12.8$, $R^2 = 0.43$, $p = 0.009$) of the total count of PPCP detections (≥ 0.005 µg/L) per site by the average total nitrogen (mg/L) per site (including only lake sites).

Survey

In order to gather information about septic systems as a possible source of PPCPs and use of the lake as a drinking water source, we surveyed lake residents about these two issues. Out of about 400 residents of the Mesalonskee Lake watershed, 33 residents responded to our survey. Survey questions and answer choices are in Appendix 2, and all survey responses are in Appendix 6. Residents who responded lived at locations all around the lakeshore (Figure 26).

Residents reported that their septic systems varied in age from less than 10 years (n=10) to between 10-20 years (n=9) to more than 20 years (n=9; Figure 26). The remaining five respondents did not know the age of their septic system. Sixteen respondents reported having their septic system inspected within the past three years, five in the last four to six

years, three within the past 12 years, four within the past 32 years, and five who did not know (Figure 27). Thus, at least 12 of the 33 respondents reported inspection times that exceed the recommended number of years between inspections (3 years; EPA 2017).

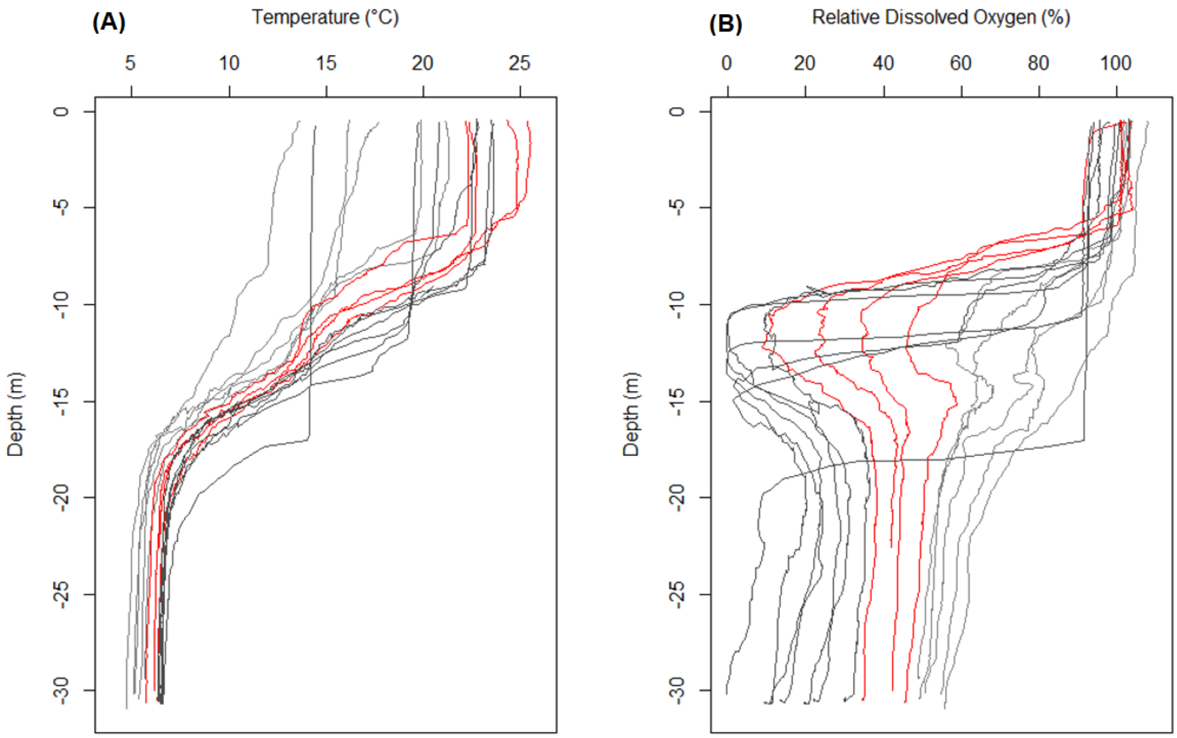

Figure 25. (A) The temperature (°C) taken across a depth profile of Messalonskee Lake in 2017. (B) The relative dissolved oxygen saturation (%) taken across a depth profile of Messalonskee Lake in 2017. Each line represents one date of temperature or dissolved oxygen readings between May 18 and November 3. Light gray lines represent dates between May 18 and June 23, 2017. The four red lines were data collected during our six weeks of sample collection, between June 30 and August 4, 2017. The dark gray lines represent data collected between August 9 and November 3, 2017.

All 25 respondents who knew the last time their septic system was pumped reported having it pumped within the past three years, which falls within the recommended amount of time (Figure 28). Ten respondents thought that septic leakage was a “very serious” problem on Messalonskee Lake, nine said “somewhat serious,” two said “not serious”, and twelve said “don’t know” (Figure 29). These responses accompany anecdotal evidence regarding the severity of this issue (Table 6).

The most common drinking water source among survey respondents was a drilled well, at 20 residents (Figure 30). Nine respondents reported using bottled water, and one respondent reported having a dug well. Only one respondent reported getting their drinking water directly from the lake, and one didn’t know what their water source was. This contradicts what we heard anecdotally from residents, that many people took their drinking water directly from the lake.

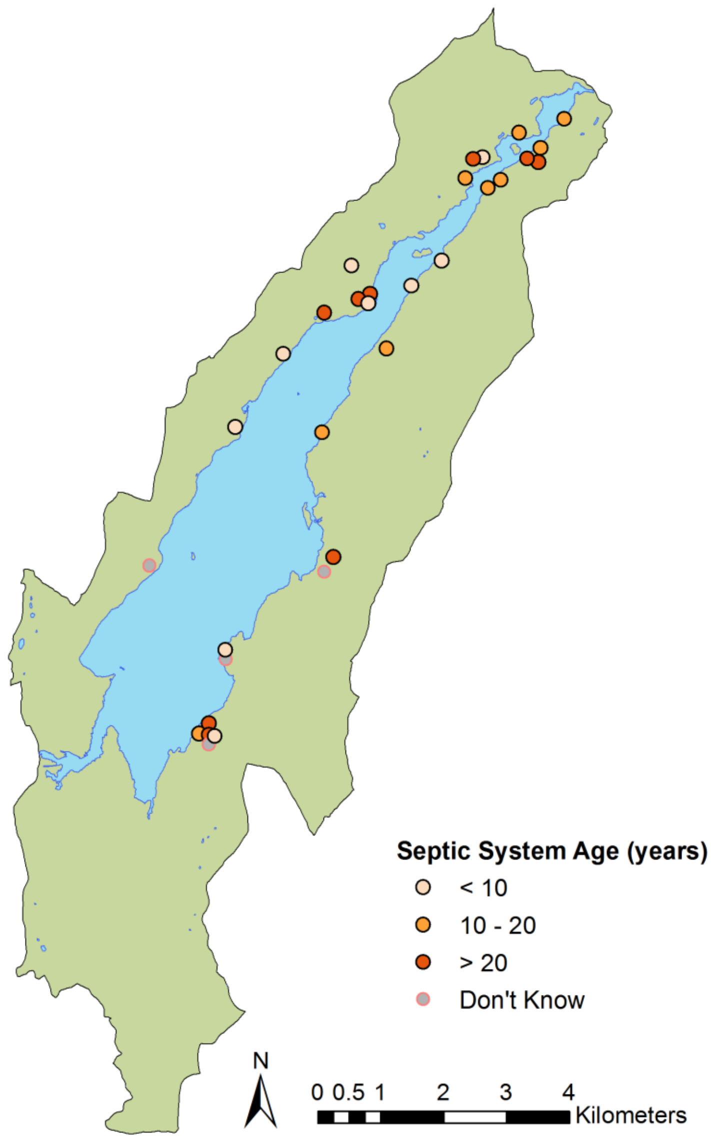

Figure 26. A map of the reported age of septic systems in the Messalonskee watershed (n = 33).

Figure 27. A map of the reported number of years since residents' septic systems have been inspected in the Messalonskee watershed (n = 33).

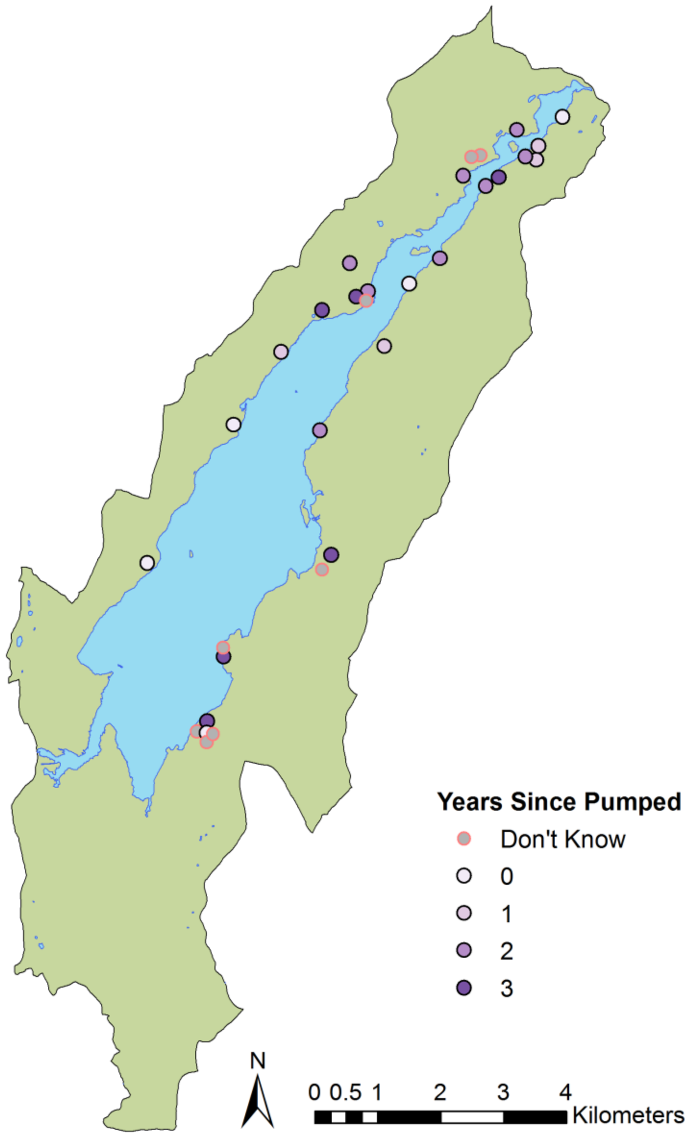

Figure 28. A map of the reported number of years since residents' septic systems have been pumped in the Messalonskee watershed (n = 33).

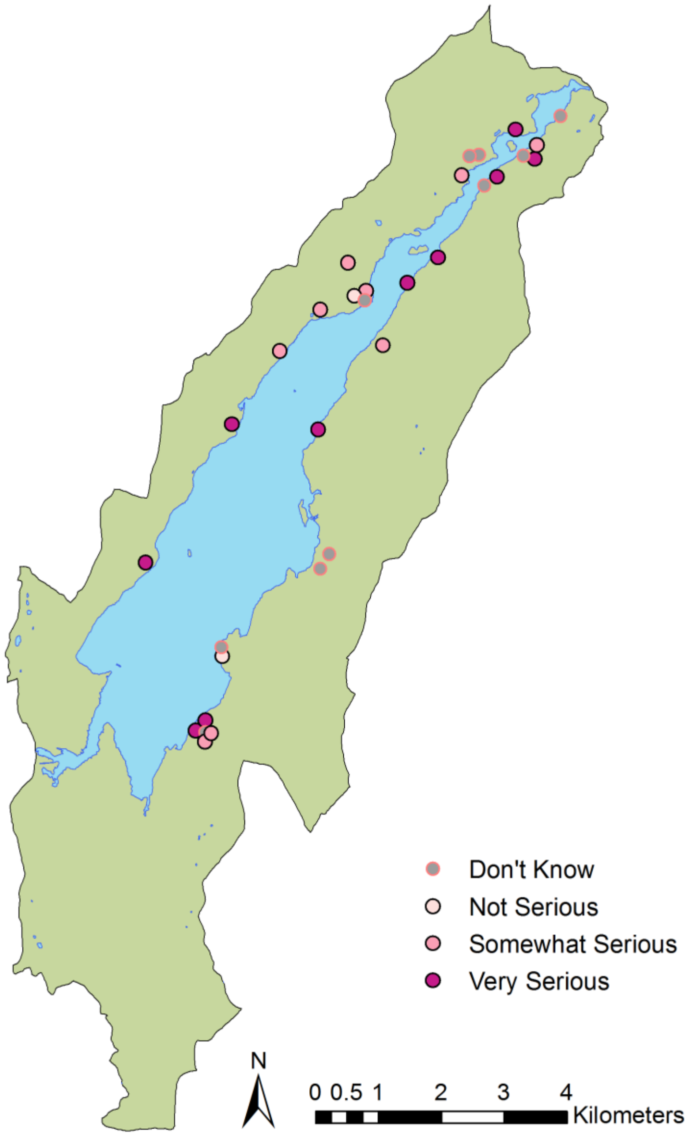

Figure 29. A map of Messalonskee residents' answers to the question "How serious of a problem do you think failing septic systems are on Messalonskee Lake?" (n = 33).

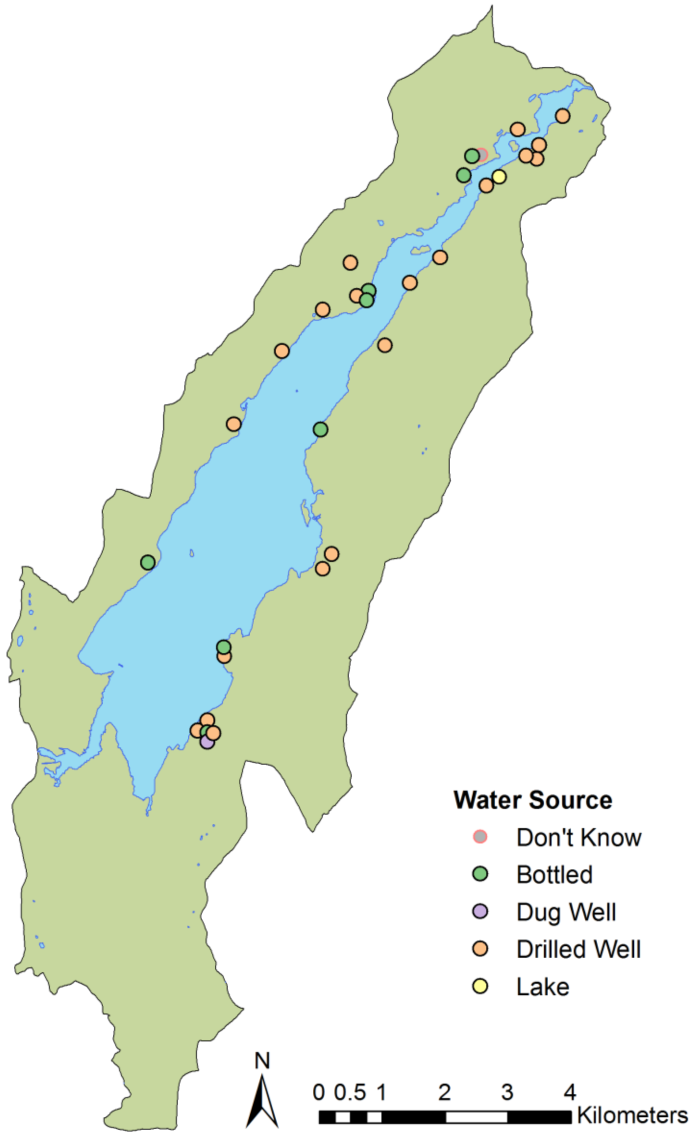

Figure 30. A map of Messalonskee residents' answers to the question "How do you obtain your drinking water?" (n = 33).

Table 6. Survey responses regarding the perceived severity of septic system failure on Messalonskee Lake, including only responses where anecdotes were provided. All responses in the second column are direct quotes.

How serious of a problem do you think failing septic systems are on Messalonskee Lake?	Please explain.
Very serious	As Messalonskee's LakeSmart spokesperson, I check home owner's septic situation and have heard about past septic problems
Don’t know	Previous house we owned had a failed system and prior to occupancy we had a whole new system installed
Somewhat serious	I’ve lived on the lake for 40 years (my entire life) and have seen first hand septics seeping into the lake.
Very serious	Check each side of me , one connected into my leach field, why I had septic guy come down, other has a barrel in the ground beside a seasonal stream!!barrel
Very serious	They have been redone in last couple of years
Somewhat serious	Neighbors commenting on a summer residence. I'm not sure if I believe it.
Somewhat serious	Just news from lake associations.
Very serious	word of mouth only
Don’t know	My landlord says, as long as you don't have a problem, don't bother.
Somewhat serious	I'm in real estate
Somewhat serious	My neighbors system failed last year, they had to dig up a section and have it pumped. Not sure if anything got in the lake.

DISCUSSION

Over four sampling dates between June 26 and August 7, 2017 and across 13 sites on Messalonskee Lake, we detected five PPCPs: caffeine, 1,7-dimethylxanthine, amphetamine, acetaminophen, and sulfachloropyridazine. Thirteen of the 18 PPCPs we tested for were not present in any of our samples. A study on PPCPs in 50 Minnesota lakes that ranged widely in human impact found an average of 3.7 PPCPs detected per lake, and a maximum of 13 PPCPs in one lake (Ferrey 2013). Our results align with those of the Minnesota study, especially for a lake that is not directly impacted by a WWTP. The concentrations of PPCPs found in our study are similar to concentrations found in other surface waters, including the Minnesota lakes study. The levels of total nitrogen detected around the lake are also in line with those found in the same lake in a previous study, with some slightly higher detections (Carey et al. 2012).

The ranges of detection for most PPCPs in this study fell between 0.005 µg/L and 0.021 µg/L. Sulfachloropyridazine fell on the very low end of this scale, with a maximum concentration of 0.007 µg/L, while amphetamine had a much more variable range that spanned from 0.005 µg/L to 0.236 µg/L. Because the total number of detections for each individual PPCP was low, most analyses depended on aggregating PPCP detections across all five chemicals. While this eliminated differences in concentration that may be important to consider in understanding PPCP dynamics in the lake, it was necessary to look for overall patterns. PPCP detection in Messalonskee Lake is associated with earlier weeks of sampling, areas of higher human use, and higher total nitrogen concentration.

Early Weeks of Sampling

It is possible that the marginal significance of the number of PPCP detections in the first week of sampling compared to the final week, which is only under a 90% significance interval, is an artifact of the presence of sulfachloropyridazine, which was present at six sites the first week of sampling (6/26/2017) and never detected thereafter. When sulfachloropyridazine detections are removed from the count, sampling week is no longer a significant factor predicting the number of PPCPs. However, sulfachloropyridazine is also present below the limit of detection at many of the sites both the first (6/26/2017) and second

weeks of sampling (7/10/2017; Appendix 4). The high detection limit, while increasing confidence of presence, may limit analysis of the dynamics of PPCPs in the lake. We acknowledge that we have fairly low count numbers of detection, which limits our analysis.

We thought that amphetamine may also be playing into the marginally significantly higher PPCP count on 6/26 versus 8/7 because it was more commonly detected compared to the other PPCPs and decreased in presence over the four weeks. Amphetamine detection by itself, however, was not significantly different across the four sampling dates. In addition, the average of the natural log of the concentration of amphetamine across the four weeks was not significantly different.

We also hypothesized that the weak pattern of decreasing PPCP presence we observed over the six weeks of sampling could be related to lake mixing dynamics. Multiple studies have found increased PPCP concentrations during winter months and decreased levels in the spring and summer (Vieno et al. 2005; Veach and Bernot 2011; Huerta-Fontela et al. 2008). Others have found that PPCP concentrations tend to be higher when water conditions are cold and flow is low due to reduced degradation in the environment (Kolpin et al. 2004). If Messalonskee Lake were still mixing or stratifying in earlier weeks of sampling, it is possible that colder water temperatures could be preventing fast degradation of PPCPs. The surface temperature during our sampling period increased significantly from the first sampling date to the following three sampling dates. This is an indication that the marginally significant decrease we saw in PPCP presence between 6/26/2017 and 8/7/2017 could be due to seasonal lake dynamics involving water temperature and degradation rates.

The presence and amount of PPCPs we observed in our study of Messalonskee Lake in the summer is comparable to the results found by Haver (2016) in three other Belgrade Lakes in the winter. We detected two PPCPs that were not detected in their study (acetaminophen and sulfachloropyridazine), and the concentrations we observed for the three PPCPs detected in both studies (caffeine, 1,7-dimethylxanthine, amphetamine) were often 2 to10-fold higher.

Many PPCPs may reach maximum concentrations in both the winter and the summer, and significant fluctuations may even be observed on a daily level for certain PPCPs, often spiking throughout and after the weekend (Huerta-Fontela et al. 2008), though these studies

have mostly been done on rivers near waste-water treatment plants. It is possible that we observed some significant concentrations of PPCPs in Messalonskee Lake in the summer due to their main mechanism of entry into the lake, whether it be direct human input or septic system leakage.

Higher Human Use

We deliberately sampled sites on Messalonskee Lake that corresponded to a range of human usage levels, including a public beach and boat launches, private residences, and open water. Our detections were significantly associated with public high-use sites compared to private sites. We chose to sample on Mondays under the assumption that if humans were a direct source of PPCPs, that weekends might be a time of greater introduction of chemicals compared to weekdays, and if the chemicals persisted for more than 24 to 48 hours, we might be more likely to detect them on Mondays. Two sites (both public high-use) in particular had more detections than the others: Oakland town beach and Belgrade boat launch (non-motorized).

It is possible that direct human input could be a major source of the PPCPs that are getting into Messalonskee Lake in the summer. Caffeine, 1,7-dimethylxanthine, and amphetamine were detected at all sites in a previous study of the Belgrade Lakes (Haver 2016), although boat launches had slightly higher levels (not statistically significant); these samples were collected in early December. It is possible that in the summer, PPCPs are detected in more localized, higher concentrations because they are being added directly to the lake through urination, direct contact with water, and dumping (Figure 31). For example, caffeine may be present in urine at 12 mg/L, six orders of magnitude higher than any of the concentrations observed in this study (Birkett and Miners 1991). Because people often carry coffee with them, it is likely that dumping coffee into the lake is another source of caffeine. In the winter, on the other hand, direct input may be less frequent, but degradation rates are slower due to lower temperatures and lower metabolic activities of microorganisms, which may allow PPCPs to spread to more areas and dilute to lower concentrations. Haver (2016) observed a similar maximum of caffeine in their study of Great Pond, East Pond, and Long

Pond (0.021 µg/L, compared to our maximum of 0.020 µg/L) and a lower average at 0.006 µg/L compared to our average of 0.011 µg/L.

Figure 31. Photos from Messalonskee Lake, Summer 2017. (A) Taken at the outlet dam (ML1); two people stand on the dam and fish, also seen drinking coffee and smoking cigarettes. (B) Taken at the Oakland public beach (ML3); parents and children enjoy a sunny day at the beach; seen swimming, eating, and drinking coffee. When asked, multiple children told us they had urinated in the lake as they were swimming. (Photos by Gail Carlson)

The Belgrade boat launch (ML11), one of the public high-use sites, is responsible for a significant amount of the PPCP detections in this human use category (GLM of PPCP detection by site type had a p-value of 0.020 if ML11 was included and 0.063 if ML11 was excluded.). The maximum concentration of PPCP from all detections was also at this site at an order of magnitude higher than at any other site (amphetamine, at 0.236 µg/L). There are a few possible explanations for why we observed more detections and higher concentrations at the Belgrade boat launch. It could be that direct input of PPCPs is higher at the Belgrade boat launch than at some other public high use sites because it is a non-motorized boat launch, meaning people must physically get in the water to get in their boats. We also observed people using this site for reasons other than to launch their boats, such as resting in their vehicles or bird watching.

It is also possible that the flow dynamics and water quality at this site have an impact on the presence and amount of PPCPs seen there. The water at this site appears to be more stagnant than in other areas of the lake, which may be attributed to the marshy surroundings with dense emergent plants. The site is below the stream inlet of the lake and in a cove, so the water flow may be slower. This means that water in this area of the lake may have a higher residence time than in other areas, preventing fast dilution of PPCPs. In addition, the dissolved oxygen concentration at the Belgrade boat launch was significantly lower than at every other site around the lake ($p < 0.0001$), averaging 2.65 ±0.10 mg/L, while the average dissolved oxygen concentration for all other lake sites was 8.47 ±0.07 mg/L. Amphetamine may be degraded in the environment through biodegradation with a half-life of about 120 hours (Bagnall et al. 2013); however, if there is not ample dissolved oxygen to support high levels of aerobic microbial activity, biodegradation rates may be lower at this site compared to others. This site is at the inlet site of the lake, which might suggest that PPCPs are flowing into the lake; however, PPCPs were only detected once in the Belgrade Stream, the inlet stream that enters the lake in a cove just north of the Belgrade boat launch. A large construction supply company and lumber yard, located between the Belgrade Stream site and the Belgrade boat launch site, creates a major land disturbance. We have no evidence that it is a source of PPCPs.

Similarly, PPCPs were only detected once at the outlet dam at the northern end of the lake, which would indicate the PPCPs that are flowing out of the lake. Most PPCPs were observationally localized; for example, the Oakland boat launch and the Oakland public beach are less than 100 meters away from each other and yet showed very different presence of PPCPs. On the other hand, when PPCPs were detected in the open water sites, they were always in at least two of the three sites at once. This observational evidence suggests the possibility that the water flow dynamics within the lake might play a large role in the movement, spread, and dilution of PPCPs in the lake.

Sulfachloropyridazine was present at many sites around the lake the first week of sampling, ≥ LOD at six sites in the northern half of the lake and below LOD but detected at almost all of the other sites. If the limit of detection had been lower (≥ 0.001 μg/L, rather than ≥0.005 μg/L in our study), sulfachloropyridazine would have been the most ubiquitous

PPCP in our panel across the first and second weeks of sampling. It is possible that sulfachloropyridazine has a higher residence time than other PPCPs and breaks down less rapidly, but so far there have been no studies (as far as we know) that have looked into breakdown methods and rates of this chemical. It is also probable that the method of entry of this PPCP into the lake would differ from other PPCPs, because sulfachloropyridazine is a veterinary antibiotic. It may enter the lake through runoff from agricultural land where livestock are kept or where manure is applied, or it may be carried by wind and deposited in the lake from agricultural or industrial sources from the lake. Indeed, certain PPCPs have been detected in the atmosphere and in precipitation (Ferrey et al. 2018). We investigated potential sources of this PPCP from the few farms in the lake watershed, but we found no evidence of usage. A large animal veterinarian in the area reported that he used very few sulfa drugs, and when they did use them, they used sulfadismethazine (pers. comm. Matt Townsend).

Septic Systems

We also hypothesized that leaking septic systems or septic field drainage may be a significant source of PPCPs entering Messalonskee Lake since no residents around the lake are connected to public wastewater treatment, and most have septic systems. Throughout the state of Maine, there are laws regarding the design, installation, and replacement of septic systems (State of Maine Department of Health and Human Services 2011). However, each town may decide the level of regulation regarding septic system inspection and maintenance. In Belgrade and Sidney it is not required that a property's septic system be inspected during a real estate transaction. There are, consequently, no comprehensive public records of septic systems for residences in those two towns. In Oakland, on the other hand, septic system inspections are required by the town, meaning that there are accurate public records of all septic systems in the town (pers. comm. Bob Ellis). The lack of consistency of records between towns, and the resulting possibility of pollution in the Messalonskee Lake watershed, may be a result of the lack of federal and state regulations on septic system inspections or a lack of property owners' compliance by with septic maintenance recommendations (Withers et al. 2014).

We found conflicting evidence about whether septic systems may be a source of PPCPs in Messalonskee Lake. We presume that private residences have septic systems, and that if septic leakage were a problem, that we would detect PPCPs at the private sampling sites. However, of the four private sampling sites across four weeks of sampling, amphetamine was the only PPCP detected, and it was only detected once at the rental property and once at the Loon Ridge Cabins at the inlet stream. Private sites are located directly downslope of septic systems and their leach fields, so the fact that we detected very little from these sites was a promising albeit small piece of evidence that septic systems around Messalonskee Lake may not be a major source. Our confidence would be higher in this regard if we had been able to sample more private sites, and if we conducted environmental sampling for direct evidence of septic failure.

We conducted a survey of residents of the Messalonskee Lake watershed to gather information on septic systems. There was a range of septic system ages among respondents, between brand new and over 20 years old, and many respondents (48%) reported having had their systems inspected within the past three years. Also, all respondents that knew when their septic system was last pumped reported that it had been done within the past three years, which is within the recommended amount of time (National Environmental Service Center 2004; EPA 2017). This evidence does not suggest that septic system leakage is a widespread issue across the watershed. We only received 33 responses to our survey; a higher number of responses would allow us more confidence in our assessment of septic health.

We also allowed residents to share their local knowledge and opinions on the potential severity of this issue. Many respondents who indicated that they believed septic leakage to be a serious issue had firsthand or secondhand accounts of problems with septic systems. This suggests that even if this issue is not widespread or a major source of PPCPs in Messalonskee Lake, it is something people are aware of. The way we framed this question may have also skewed the results. Allowing respondents to say they were unsure as to the severity of septic leakage may have decreased the number of responses we saw for “not serious” because respondents could have answered “don’t know” if they had never heard of any cases of septic leakage being a problem. Another potential issue with this question is that

simply asking it might imply to respondents that it is an issue, and they may therefore be more likely to say that it is indeed a serious problem. Gathering respondents' descriptions of what they know about the problem helps us gather more useful data. Likewise, residents may have been more likely to respond to the survey in the first place if this is an issue they already care about and if they are more concerned residents that take better care of their property.

Lastly, we looked into correlations between PPCP detections and total nitrogen and total phosphorus, since higher levels of these nutrients may come from septic systems and/or be a general indicator of water quality at each of our sampling sites. It was expected that total phosphorus would not correlate to total nitrogen or PPCP detections because Messalonskee Lake, like many aquatic systems, is phosphorus-limited. This means that excess phosphorus is typically taken up quickly by phytoplankton. However, the fact that PPCP detection did correlate significantly to total nitrogen levels by site does indicate the possibility that septic system leakage could be contributing these PPCPs to the lake if septic systems are a major source of nitrogen on this lake. On the other hand, yard and agricultural runoff can also lead to elevated levels of total nitrogen, so there is also considerable uncertainty in this explanation.

Ecological and Human Health Impacts

The PPCPs that were detected in Messalonskee Lake may be of ecological and human health concern, although these potential impacts were not explored in this study. That said, the typically low concentrations detected and seemingly short-lived nature of PPCPs in Messalonskee Lake indicates that human health consequences may not be of particularly high concern. Only one survey respondent out of 33 reported taking their drinking water directly from the lake, and they utilize a complex filtering mechanism that may be effective at removing some PPCPs. It is possible that sulfachloropyridazine, a veterinary antibiotic, could promote antibiotic resistance in lake microbes, and could affect microbial communities in the long-term. Long-term ingestion of acetaminophen may not be harmful to human health at low levels (Schwab et al. 2005), but ecological impacts, especially in combination with other PPCPs like caffeine, may be significant (Fraker and Smith 2004). Exposure to caffeine may

affect the activity levels of many aquatic organisms (Fraker and Smith 2004; Crane et al. 2006). Amphetamine's impacts on human health are well-known, as amphetamine is a pharmaceutical substance found in ADHD and weight-loss medication. Its effects from exposure to low levels in surface water are unknown, but its ecological impacts may be serious, including oxidative and genetic impacts on zebra mussels (Parolini et al. 2016).

Future studies should investigate how the levels and combinations of PPCPs found in Messalonskee Lake may affect long-term ecological and human health. Analytical methods for detecting caffeine in water are less expensive, for example, meaning higher temporal and spatial resolution in data may be achieved, allowing further insight into the dynamics of PPCPs in Messalonskee Lake. A single 12 oz. cup of brewed coffee from Starbucks has on average 233 mg caffeine. If one cup of coffee is dissolved into Messalonskee Lake, which has a volume of about 1.36×10^{11} L of water, the resulting concentration of caffeine would be 0.0017 ng/L, which is three to four orders of magnitude lower than what we observed at the sites where caffeine was detected. Since the lake is stratified in the summer, preventing mixing between the epilimnion and hypolimnion, the caffeine from one cup of coffee would not mix to the deeper layers of the lake. Also, lake flow dynamics may prevent it from quickly dissolving across the surface of the lake as well, which may be why we observed higher concentrations in localized areas.

Changes across the season may be due to changes in levels of UV radiation or biotic activity, which both may be measured alongside PPCPs in the future to try to further explore and explain seasonal dynamics. Additionally, to gain a deeper understanding of the short-term dynamics of PPCPs in the lake, taking samples across multiple times per week may provide more detailed insight on the residence time of different PCPPs.

Conclusion

Messalonskee Lake is not a highly contaminated system regarding PPCP contamination. We only detected five chemicals, the total number of detections was fairly low, and the number of detections decreased marginally over time. It is also similar to other lakes that are not impacted by WWTPs or are in remote locations (Ferrey 2013; Ferrey 2015). However, two of our high use sites were where most PPCPs are found—one likely

due to direct human contact with the water, and the other likely due to being highly degraded—so high use sites may be worth future attention. Our study was not able to confirm sources of PPCPs in Messalonskee Lake.

Given that nearly every lake that has been studied has low levels of some PPCPs, it may be an inevitable category of contaminant in surface waters. Human usage of lakes may unavoidably lead to this type of contamination. We should be less concerned with how to completely prevent PPCP contamination in the environment and focus more on how we might be able to mitigate the potential ecological and human health impacts that these chemicals pose, especially in localized sites of high impact. This study represents one of two that have been conducted looking at PPCPs in Maine lakes, meaning a large knowledge gap remains in terms of describing PPCP presence in the state's surface waters. Messalonskee Lake may be used as a model system for exploring the potentially low levels of PPCP presence and impact in similar lakes across the state.

LITERATURE CITED

Andreozzi, R., M. Raffaele, and P. Nicklas. 2003. Pharmaceuticals in STP effluents and their solar photodegradation in aquatic environment. Chemosphere **50**:1319-1330.

Bacon, L. 2013. Water quality in the Belgrade Lakes. http://belgradelakesassociation.org/Portals/0/PDFs/Resources%20Water%20Quality/Linda%20Bacon.pdf

Bagnall, J., L. Malia, A. Lubben, and B. Kasprzyk-Hordern. 2013. Stereoselective biodegradation of amphetamine and methamphetamine in river microcosms. Water Research **47**:5708-5718.

Baker, D. R., and B. Kasprzyk-Hordern. 2013. Spatial and temporal occurrence of pharmaceuticals and illicit drugs in the aqueous environment and during wastewater treatment: new developments. Science of The Total Environment **454**:442-456.

Bartelt-Hunt, S. L., D. D. Snow, T. Damon, J. Shockley, and K. Hoagland. 2009. The occurrence of illicit and therapeutic pharmaceuticals in wastewater effluent and surface waters in Nebraska. Environmental Pollution **157**:786-791.

Berninger, J. P., B. Du, K. A. Connors, S. A. Eytcheson, M. A. Kolkmeier, K. N. Prosser, T. W. Valenti, C. K. Chambliss, and B. W. Brooks. 2011. Effects of the antihistamine diphenhydramine on selected aquatic organisms. Environmental Toxicology and Chemistry **30**:2065-2072.

Birkett, D., and J. Miners. 1991. Caffeine renal clearance and urine caffeine concentrations during steady state dosing. Implications for monitoring caffeine intake during sports events. British journal of clinical pharmacology **31**:405-408.

Boles, T. H., and M. J. Wells. 2010. Analysis of amphetamine and methamphetamine as emerging pollutants in wastewater and wastewater-impacted streams. Journal of Chromatography A **1217**:2561-2568.

Boxall, A. B., M. A. Rudd, B. W. Brooks, D. J. Caldwell, K. Choi, S. Hickmann, E. Innes, K. Ostapyk, J. P. Staveley, and T. Verslycke. 2012. Pharmaceuticals and personal care products in the environment: what are the big questions? Environmental health perspectives **120**:1221.

Buerge, I. J., M. Kahle, H.-R. Buser, M. D. Müller, and T. Poiger. 2008. Nicotine derivatives in wastewater and surface waters: application as chemical markers for domestic wastewater. Environmental science & technology **42**:6354-6360.

Burgess, I., and M. K. Nelson. 2009. Belgrade lakes Maine: the history of a summer community and its effect on the environment. http://belgradelakesassociation.org/Portals/0/PDFs/General/Belgrade%20Lakes%20History1.pdf

Buth, J. M., W. A. Arnold, and K. McNeill. 2007. Unexpected products and reaction mechanisms of the aqueous chlorination of cimetidine. Environmental science & technology **41**:6228-6233.

Carey, C. C., H. A. Ewing, K. L. Cottingham, K. C. Weathers, R. Q. Thomas, and J. F. Haney. 2012. Occurrence and toxicity of the cyanobacterium *Gloeotrichia echinulata* in low-nutrient lakes in the northeastern United States. Aquatic Ecology 46:395-409.

Castillo, L. E., E. de la Cruz, and C. Ruepert. 1997. Ecotoxicology and pesticides in tropical aquatic ecosystems of Central America. Environmental Toxicology and Chemistry **16**:41-51.

(CDC) Centers for Disease Control and Prevention. 2015. Community water treatment. https://www.cdc.gov/healthywater/drinking/public/water_treatment.html

Center for Sustainable Systems, University of Michigan. 2016. U.S. Wastewater treatment factsheet. Pub. No. CSS04-14. http://css.umich.edu/sites/default/files/U.S._Wastewater_Treatment_Factsheet_CSS04-14.pdf

Choi, K., Y. Kim, J. Jung, M. H. Kim, C. S. Kim, N. H. Kim, and J. Park. 2008. Occurrences and ecological risks of roxithromycin, trimethoprim, and chloramphenicol in the Han River, Korea. Environmental Toxicology and Chemistry **27**:711-719.

Clara, M., B. Strenn, and N. Kreuzinger. 2004. Carbamazepine as a possible anthropogenic marker in the aquatic environment: investigations on the behaviour of carbamazepine in wastewater treatment and during groundwater infiltration. Water Research **38**:947-954.

Colby Environmental Assessment Team. 1997. Land use patterns in relation to lake water quality in the Messalonskee Lake watershed. Colby College, Waterville, ME. http://digitalcommons.colby.edu/cgi/viewcontent.cgi?article=1001&context=otherlakes

Corcoran, J., M. J. Winter, and C. R. Tyler. 2010. Pharmaceuticals in the aquatic environment: a critical review of the evidence for health effects in fish. Critical reviews in toxicology **40**:287-304.

Crane, M., C. Watts, and T. Boucard. 2006. Chronic aquatic environmental risks from exposure to human pharmaceuticals. Science of The Total Environment **367**:23-41.

De la Torre, R., M. Farré, P. N. Roset, N. Pizarro, S. Abanades, M. Segura, J. Segura, and J. Camí. 2004. Human pharmacology of MDMA: pharmacokinetics, metabolism, and disposition. Therapeutic drug monitoring **26**:137-144.

Deo, R. P. 2014. Pharmaceuticals in the surface water of the USA: A review. Current Environmental Health Reports **1**:113-122.

EPA (US Environmental Protection Agency). 2005. A homeowner's guide to septic systems. https://www3.epa.gov/npdes/pubs/homeowner_guide_long.pdf

EPA (US Environmental Protection Agency). 2016a. Municipal wastewater. https://www.epa.gov/npdes/municipal-wastewater

EPA (US Environmental Protection Agency). 2016b. Secondary treatment standards. https://www.epa.gov/npdes/secondary-treatment-standards

EPA (US Environmental Protection Agency). 2016c. About private water wells. https://19january2017snapshot.epa.gov/privatewells/about-private-water-wells_.html

EPA (US Environmental Protection Agency). 2017. How to care for your septic system. https://www.epa.gov/septic/how-care-your-septic-system

ESRI. 2011. ArcGIS Desktop: Release 10. Redlands, CA: Environmental Systems Research Institute.

Fairbairn, D. J., M. E. Karpuzcu, W. A. Arnold, B. L. Barber, E. F. Kaufenberg, W. C. Koskinen, P. J. Novak, P. J. Rice, and D. L. Swackhamer. 2016. Sources and transport of contaminants of emerging concern: a two-year study of occurrence and spatiotemporal variation in a mixed land use watershed. Science of The Total Environment **551**:605-613.

Farré, M. l., S. Pérez, L. Kantiani, and D. Barceló. 2008. Fate and toxicity of emerging pollutants, their metabolites and transformation products in the aquatic environment. TrAC Trends in Analytical Chemistry **27**:991-1007.

Fenz, R., A. P. Blaschke, M. Clara, H. Kroiss, D. Mascher, and M. Zessner. 2005. Monitoring of carbamazepine concentrations in wastewater and groundwater to quantify sewer leakage. Water science and technology **52**:205-213.

Ferrey, M. 2013. Pharmaceuticals and endocrine active chemicals in Minnesota lakes. Minnesota Pollution Control Agency, Saint Paul, MN. https://www.pca.state.mn.us/sites/default/files/tdr-g1-16.pdf

Ferrey, M. 2015. Pharmaceuticals, personal care products, and endocrine active chemical monitoring in lakes and rivers: 2013. Minnesota Pollution Control Agency. Saint Paul, MN. https://www.pca.state.mn.us/sites/default/files/tdr-g1-18.pdf

Ferrey, M. L., M. C. Hamilton, W. J. Backe, and K. E. Anderson. 2018. Pharmaceuticals and other anthropogenic chemicals in atmospheric particulates and precipitation. Science of The Total Environment **612**:1488-1497.

Fleck, H. n. d. How to filter lake water for residential use. Culligan. http://www.culligan.com/home/solution-center/resources/how-to-filter-lake-water-for-residential-use

Fraker, S. L., and G. R. Smith. 2004. Direct and interactive effects of ecologically relevant concentrations of organic wastewater contaminants on Rana pipiens tadpoles. Environmental toxicology **19**:250-256.

Friends of Messalonskee.2017. Milfloil control. http://www.friendsofmessalonskee.com/milfoil_control

Gagné, F., C. Blaise, M. Fournier, and P. Hansen. 2006. Effects of selected pharmaceutical products on phagocytic activity in Elliptio complanata mussels. Comparative Biochemistry and Physiology Part C: Toxicology & Pharmacology **143**:179-186.

Gao, P., Y. Ding, H. Li, and I. Xagoraraki. 2012. Occurrence of pharmaceuticals in a municipal wastewater treatment plant: mass balance and removal processes. Chemosphere **88**:17-24.

Goolsby, E. W., C. M. Mason, J. T. Wojcik, A. M. Jordan, and M. C. Black. 2013. Acute and chronic effects of diphenhydramine and sertraline mixtures in Ceriodaphnia dubia. Environmental Toxicology and Chemistry **32**:2866-2869.

Haver, S. 2016. Pharmaceutical and personal care products concentrations in the Belgrade Lakes: A possible threat to aquatic ecosystems and human health. Colby College Honors Theses: Paper 810. http://digitalcommons.colby.edu/honorstheses/810

Hedgespeth, M. L., Y. Sapozhnikova, P. Pennington, A. Clum, A. Fairey, and E. Wirth. 2012. Pharmaceuticals and personal care products (PPCPs) in treated wastewater discharges into Charleston Harbor, South Carolina. Science of The Total Environment **437**:1-9.

Hoppe, P. D., E. J. Rosi-Marshall, and H. A. Bechtold. 2012. The antihistamine cimetidine alters invertebrate growth and population dynamics in artificial streams. Freshwater science **31**:379-388.

Huber, D., K. Bleymaier, A. Pellis, R. Vielnascher, A. Daxbacher, K. J. Greimel, and G. M. Guebitz. 2018. Laccase catalyzed elimination of morphine from aqueous systems. New biotechnology.

Huerta-Fontela, M., M. T. Galceran, and F. Ventura. 2008. Stimulatory drugs of abuse in surface waters and their removal in a conventional drinking water treatment plant. Environmental science & technology **42**:6809-6816.

Ji, K., S. Kim, S. Han, J. Seo, S. Lee, Y. Park, K. Choi, Y.-L. Kho, P.-G. Kim, and J. Park. 2012. Risk assessment of chlortetracycline, oxytetracycline, sulfamethazine, sulfathiazole, and erythromycin in aquatic environment: are the current environmental concentrations safe? Ecotoxicology **21**:2031-2050.

Jung, J., Y. Kim, J. Kim, D.-H. Jeong, and K. Choi. 2008. Environmental levels of ultraviolet light potentiate the toxicity of sulfonamide antibiotics in Daphnia magna. Ecotoxicology **17**:37-45.

Karthikeyan, K., and M. T. Meyer. 2006. Occurrence of antibiotics in wastewater treatment facilities in Wisconsin, USA. Science of The Total Environment **361**:196-207.

Kasprzyk-Hordern, B., R. M. Dinsdale, and A. J. Guwy. 2009. The removal of pharmaceuticals, personal care products, endocrine disruptors and illicit drugs during wastewater treatment and its impact on the quality of receiving waters. Water Research **43**:363-380.

Kawabata, K., K. Sugihara, S. Sanoh, S. Kitamura, and S. Ohta. 2012. Ultraviolet-photoproduct of acetaminophen: Structure determination and evaluation of ecotoxicological effect. Journal of Photochemistry and Photobiology A: Chemistry **249**:29-35.

Kennedy, C. D., S. W. Houmes, K. L. Wyrick, S. M. Kammerzell, K. Lukowiak, and B. A. Sorg. 2010. Methamphetamine enhances memory of operantly conditioned respiratory behavior in the snail Lymnaea stagnalis. Journal of Experimental Biology **213**:2055-2065.

Kim, S., and D. S. Aga. 2007. Potential ecological and human health impacts of antibiotics and antibiotic-resistant bacteria from wastewater treatment plants. Journal of Toxicology and Environmental Health, Part B **10**:559-573.

Kim, Y., K. Choi, J. Jung, S. Park, P.-G. Kim, and J. Park. 2007. Aquatic toxicity of acetaminophen, carbamazepine, cimetidine, diltiazem and six major sulfonamides, and their potential ecological risks in Korea. Environment International **33**:370-375.

Kolpin, D. W., E. T. Furlong, M. T. Meyer, E. M. Thurman, S. D. Zaugg, L. B. Barber, and H. T. Buxton. 2002. Pharmaceuticals, hormones, and other organic wastewater contaminants in US streams, 1999− 2000: A national reconnaissance. Environmental science & technology **36**:1202-1211.

Kolpin, D. W., M. Skopec, M. T. Meyer, E. T. Furlong, and S. D. Zaugg. 2004. Urban contribution of pharmaceuticals and other organic wastewater contaminants to streams during differing flow conditions. Science of The Total Environment **328**:119-130.

Kolpin, D. W. 2017. Contaminants of emerging concern in the environment. U.S. Geological Survey. https://toxics.usgs.gov/investigations/cec/index.php

Kozuskanich, J. C., K. S. Novakowski, B. C. Anderson, A. S. Crowe, and V. K. Balakrishnan. 2013. Anthropogenic impacts on a bedrock aquifer at the village scale. Groundwater **52**:474-486.

Lam, M. W., C. J. Young, R. A. Brain, D. J. Johnson, M. A. Hanson, C. J. Wilson, S. M. Richards, K. R. Solomon, and S. A. Mabury. 2004. Aquatic persistence of eight pharmaceuticals in a microcosm study. Environmental Toxicology and Chemistry **23**:1431-1440.

Latch, D. E., J. L. Packer, W. A. Arnold, and K. McNeill. 2003. Photochemical conversion of triclosan to 2, 8-dichlorodibenzo-p-dioxin in aqueous solution. Journal of Photochemistry and Photobiology A: Chemistry **158**:63-66.

Lee, S., D. Jung, Y. Kho, K. Ji, P. Kim, B. Ahn, and K. Choi. 2015. Ecotoxicological assessment of cimetidine and determination of its potential for endocrine disruption using three test organisms: Daphnia magna, Moina macrocopa, and Danio rerio. Chemosphere **135**:208-216.

Lin, A. Y.-C., C.-A. Lin, H.-H. Tung, and N. S. Chary. 2010. Potential for biodegradation and sorption of acetaminophen, caffeine, propranolol and acebutolol in lab-scale aqueous environments. Journal of Hazardous Materials **183**:242-250.

Lopes, A., N. Silva, M. Bronze, J. Ferreira, and J. Morais. 2014. Analysis of cocaine and nicotine metabolites in wastewater by liquid chromatography–tandem mass spectrometry. Cross abuse index patterns on a major community. Science of The Total Environment **487**:673-680.

Maine State Legislature. 2013. Title 38: Waters and navigation: Chapter 3: Protection and improvement of waters: Subchapter 1: Environmental Protection Board: Article 4-A: Water Classification Program. http://legislature.maine.gov/statutes/38/title38sec465-A.html

Maine Volunteer Lake Monitoring Program. 2018. Messalonskee Lake (Snow Pond). https://www.lakesofmaine.org/lake-overview.html?m=5280

Malarvizhi, A., C. Kavitha, M. Saravanan, and M. Ramesh. 2012. Carbamazepine (CBZ) induced enzymatic stress in gill, liver and muscle of a common carp, Cyprinus carpio. Journal of King Saud University-Science **24**:179-186.

ME DEP. n.d. Lakes. Lakes. http://www.maine.gov/dep/water/lakes/

Meidel, S. K. 2016. Classification of Maine waters. http://www.maine.gov/dep/water/monitoring/classification/

Metcalfe, C., K. Tindale, H. Li, A. Rodayan, and V. Yargeau. 2010. Illicit drugs in Canadian municipal wastewater and estimates of community drug use. Environmental Pollution **158**:3179-3185.

Michael, I., L. Rizzo, C. McArdell, C. Manaia, C. Merlin, T. Schwartz, C. Dagot, and D. Fatta-Kassinos. 2013. Urban wastewater treatment plants as hotspots for the release of antibiotics in the environment: a review. Water Research **47**:957-995.

Nassef, M., S. Matsumoto, M. Seki, F. Khalil, I. J. Kang, Y. Shimasaki, Y. Oshima, and T. Honjo. 2010. Acute effects of triclosan, diclofenac and carbamazepine on feeding performance of Japanese medaka fish (Oryzias latipes). Chemosphere **80**:1095-1100.

National Environmental Services Center. 2004. Maintaining your septic system--A guide for homeowners. Pipeline (15). Retrieved from http://www.nesc.wvu.edu/pdf/ww/septic/pl_fall04.pdf

National Environmental Services Center. 2007. Pharmaceuticals and personal care products: An overview. Pipeline (18). http://www.nesc.wvu.edu/pdf/WW/publications/pipline/pl_wi07.pdf

National Research Council. 1996. Municipal wastewater and sludge treatment. Use of reclaimed water and sludge in food crop production. Washington, DC: The National Academies Press. https://www3.epa.gov/npdes/pubs/mstr-ch3.pdf

Nikolaou, A., S. Meric, and D. Fatta. 2007. Occurrence patterns of pharmaceuticals in water and wastewater environments. Analytical and bioanalytical chemistry **387**:1225-1234.

O'Hara, F. n.d. Septic systems: How they work and how to keep them going. Maine Department of Environmental Protection. http://www.maine.gov/dep/water/groundwater/septic_systems.pdf on June 14, 2017

Parolini, M., S. Magni, S. Castiglioni, and A. Binelli. 2016. Amphetamine exposure imbalanced antioxidant activity in the bivalve Dreissena polymorpha causing oxidative and genetic damage. Chemosphere **144**:207-213.

Pastrana-Martínez, L. M., J. L. Faria, J. M. Doña-Rodríguez, C. Fernández-Rodríguez, and A. M. T. Silva. 2012. Degradation of diphenhydramine pharmaceutical in aqueous solutions by using two highly active TiO2 photocatalysts: Operating parameters and photocatalytic mechanism. Applied Catalysis B: Environmental **113-114**:221-227.

Pease, A. M., H. M. Krumholz, N. S. Downing, J. A. Aminawung, N. D. Shah, and J. S. Ross. 2017. Postapproval studies of drugs initially approved by the FDA on the basis of limited evidence: systematic review. bmj **357**:j1680.

Peréz, J. S., I. Carra, C. Sirtori, A. Agüera, and B. Esteban. 2014. Fate of thiabendazole through the treatment of a simulated agro-food industrial effluent by combined MBR/Fenton processes at μg/L scale. Water Research **51**:55-63.

Pérez, S., P. Eichhorn, and D. S. Aga. 2005. Evaluating the biodegradability of sulfamethazine, sulfamethoxazole, sulfathiazole, and trimethoprim at different stages of sewage treatment. Environmental Toxicology and Chemistry **24**:1361-1367.

Petrie, B., R. Barden, and B. Kasprzyk-Hordern. 2015. A review on emerging contaminants in wastewaters and the environment: current knowledge, understudied areas and recommendations for future monitoring. Water Research **72**:3-27.

Petrović, M., S. Gonzalez, and D. Barceló. 2003. Analysis and removal of emerging contaminants in wastewater and drinking water. TrAC Trends in Analytical Chemistry **22**:685-696.

Pieper, C., D. Risse, B. Schmidt, B. Braun, U. Szewzyk, and W. Rotard. 2010. Investigation of the microbial degradation of phenazone-type drugs and their metabolites by natural biofilms derived from river water using liquid chromatography/tandem mass spectrometry (LC-MS/MS). Water Research **44**:4559-4569.

Pomati, F., S. Castiglioni, E. Zuccato, R. Fanelli, D. Vigetti, C. Rossetti, and D. Calamari. 2006. Effects of a complex mixture of therapeutic drugs at environmental levels on human embryonic cells. Environmental science & technology **40**:2442-2447.

Pomati, F., C. Orlandi, M. Clerici, F. Luciani, and E. Zuccato. 2007. Effects and interactions in an environmentally relevant mixture of pharmaceuticals. Toxicological Sciences **102**:129-137.

Qualtrics. 2018. Qualtrics (June 2017). Qualtrics, Provo, UT. https://www.qualtrics.com

Reed, E., and S. Haver. 2015. Septic system influence on eutrophication: Are septic systems a significant source of nutrients to the Belgrade Lakes?. Belgrade Lakes water quality report: Problems, consequences, and solutions. Colby College, Waterville, ME.

Richmond, E. K., M. R. Grace, J. J. Kelly, A. J. Reisinger, E. J. Rosi, and D. M. Walters. 2017. Pharmaceuticals and personal care products (PPCPs) are ecological disrupting compounds (EcoDC). Elem Sci Anth **5**.

Rosi-Marshall, E. J., D. Snow, S. L. Bartelt-Hunt, A. Paspalof, and J. L. Tank. 2015. A review of ecological effects and environmental fate of illicit drugs in aquatic ecosystems. Journal of Hazardous Materials **282**:18-25.

RStudio Team 2015. RStudio: Integrated development for R (v. 3.4.2). RStudio, Inc., Boston, MA. http://www.rstudio.com/

Ryan, C. C., D. T. Tan, and W. A. Arnold. 2011. Direct and indirect photolysis of sulfamethoxazole and trimethoprim in wastewater treatment plant effluent. Water Research **45**:1280-1286.

Sanderson, H., D. J. Johnson, C. J. Wilson, R. A. Brain, and K. R. Solomon. 2003. Probabilistic hazard assessment of environmentally occurring pharmaceuticals toxicity to fish, daphnids and algae by ECOSAR screening. Toxicology letters **144**:383-395.

Schmitt, H., P. Van Beelen, J. Tolls, and C. L. Van Leeuwen. 2004. Pollution-induced community tolerance of soil microbial communities caused by the antibiotic sulfachloropyridazine. Environmental science & technology **38**:1148-1153.

Schnell, S., N. C. Bols, C. Barata, and C. Porte. 2009. Single and combined toxicity of pharmaceuticals and personal care products (PPCPs) on the rainbow trout liver cell line RTL-W1. Aquatic Toxicology **93**:244-252.

Schriks, M., M. B. Heringa, M. M. van der Kooi, P. de Voogt, and A. P. van Wezel. 2010. Toxicological relevance of emerging contaminants for drinking water quality. Water Research **44**:461-476.

Schwab, B. W., E. P. Hayes, J. M. Fiori, F. J. Mastrocco, N. M. Roden, D. Cragin, R. D. Meyerhoff, J. Vincent, and P. D. Anderson. 2005. Human pharmaceuticals in US surface waters: a human health risk assessment. Regulatory Toxicology and Pharmacology **42**:296-312.

Stackelberg, P. E., J. Gibs, E. T. Furlong, M. T. Meyer, S. D. Zaugg, and R. L. Lippincott. 2007. Efficiency of conventional drinking-water-treatment processes in removal of pharmaceuticals and other organic compounds. Science of The Total Environment **377**:255-272.

State of Maine Department of Health and Human Services. 2011. Subsurface wastewater disposal rules. 10-144 CMR 241. https://www1.maine.gov/dhhs/mecdc/environmental-health/plumb/documents/pdf/10-144-cmr-241-final.pdf

Tarpani, R. R. Z., and A. Azapagic. 2018. A methodology for estimating concentrations of pharmaceuticals and personal care products (PPCPs) in wastewater treatment plants and in freshwaters. Science of The Total Environment **622-623**:1417-1430.

Trovó, A. G., R. F. Nogueira, A. Agüera, C. Sirtori, and A. R. Fernández-Alba. 2009. Photodegradation of sulfamethoxazole in various aqueous media: persistence, toxicity and photoproducts assessment. Chemosphere **77**:1292-1298.

Veach, A. M., and M. J. Bernot. 2011. Temporal variation of pharmaceuticals in an urban and agriculturally influenced stream. Science of The Total Environment **409**:4553-4563.

Verlicchi, P., M. Al Aukidy, and E. Zambello. 2012. Occurrence of pharmaceutical compounds in urban wastewater: removal, mass load and environmental risk after a secondary treatment—a review. Science of The Total Environment **429**:123-155.

Verstraeten, I. M., G. S. Fetterman, S. K. Sebree, M. T. Meyer, and T. D. Bullen. 2004. Is septic waste affecting drinking water from shallow domestic wells along the Platte River in eastern Nebraska. USGS Fact Sheet:072-003.

Vieno, N. M., T. Tuhkanen, and L. Kronberg. 2005. Seasonal variation in the occurrence of pharmaceuticals in effluents from a sewage treatment plant and in the recipient water. Environmental science & technology **39**:8220-8226.

WHO (World Health Organization). 2012. Pharmaceuticals in drinking-water. France. http://apps.who.int/iris/bitstream/10665/44630/1/9789241502085_eng.pdf

Wick, A., G. Fink, A. Joss, H. Siegrist, and T. A. Ternes. 2009. Fate of beta blockers and psycho-active drugs in conventional wastewater treatment. Water Research **43**:1060-1074.

Wilson, B. A., V. H. Smith, F. deNoyelles, and C. K. Larive. 2003. Effects of three pharmaceutical and personal care products on natural freshwater algal assemblages. Environmental science & technology **37**:1713-1719.

Withers, P. J., P. Jordan, L. May, H. P. Jarvie, and N. E. Deal. 2014. Do septic tank systems pose a hidden threat to water quality? Frontiers in Ecology and the Environment **12**:123-130.

Yamamoto, H., Y. Nakamura, S. Moriguchi, Y. Nakamura, Y. Honda, I. Tamura, Y. Hirata, A. Hayashi, and J. Sekizawa. 2009. Persistence and partitioning of eight selected pharmaceuticals in the aquatic environment: laboratory photolysis, biodegradation, and sorption experiments. Water Research **43**:351-362.

Yang, S., J. Cha, and K. Carlson. 2005. Simultaneous extraction and analysis of 11 tetracycline and sulfonamide antibiotics in influent and effluent domestic wastewater by solid-phase extraction and liquid chromatography-electrospray ionization tandem mass spectrometry. Journal of Chromatography A **1097**:40-53.

Yang, Y., Y. S. Ok, K.-H. Kim, E. E. Kwon, and Y. F. Tsang. 2017. Occurrences and removal of pharmaceuticals and personal care products (PPCPs) in drinking water and water/sewage treatment plants: A review. Science of The Total Environment **596-597**:303-320.

Zuccato, E., and S. Castiglioni. 2009. Illicit drugs in the environment. Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences **367**:3965-3978.

Zühlke, S., U. Dünnbier, and T. Heberer. 2004. Detection and identification of phenazone-type drugs and their microbial metabolites in ground and drinking water applying solid-phase extraction and gas chromatography with mass spectrometric detection. Journal of Chromatography A **1050**:201-209.

APPENDIX 1. Exact locations of Messalonskee Lake, Kennebec River, and Messalonskee Stream sampling sites.

Site ID	Site description	Substrate	Latitude	Longitude
ML1	Outlet dam	Gravel, concrete, detritus	44.540055	-69.723163
ML2	Oakland boat launch	Concrete ramp, rip-rap	44.540209	-69.726102
ML3	Oakland beach	Gravel, leaf litter	44.540504	-69.727241
ML4	North Buoy (MESSDEP 2)	Deep water	44.519510	-69.750590
ML5	Middle of South Buoy (MESSDEP 1)	Deep water	44.497740	-69.774210
ML6	Middle of lake	Deep water	44.487408	-69.786192
ML7	Private residence (East)	Pebbles, muck, rock	44.525600	-69.744032
ML8	Hume Center	Sand, rocks, boulders	44.510067	-69.759537
ML9	Sidney boat launch	Concrete ramp, rip-rap	44.490754	-69.776670
ML10	Rental property	Boulders, rocks	44.460372	-69.794611
ML11	Belgrade boat launch	Concrete ramp, muck, milfoil	44.442590	-69.830790
ML12	Inlet stream (Loon Ridge Cabins)	Rocks, milfoil	44.441866	-69.832243
ML13	Private residence (West)	Sand, rocks	44.491503	-69.791529
KR1	River, downstream of Kennebec Sanitary Treatment plant	Rocky, fast current	44.524461	-69.656632
KR2	River, upstream of Kennebec Sanitary Treatment plant	Concrete, rocks	44.533175	-69.644094
MS	North St. boat launch	Gravel, few big rocks	44.564622	-69.648747

APPENDIX 2. The following is the text-version of the online survey taken by residents of the Messalonskee Watershed:

Messalonskee Lake Residents Survey on Drinking Water and Septic Systems

My name is Alyssa Kullberg, and I am a Colby College student conducting a research project on water quality on Messalonskee Lake. My research advisor is Professor Gail Carlson, and we are collaborating with the Friends of Messalonskee Watershed.

I would like to gather information about drinking water and septic systems on Messalonskee Lake. Please kindly take a few minutes to fill out this survey about your drinking water and septic system. All information will be kept anonymous; we will not collect names and no information will be traceable to specific survey participants.

Consent Form
Colby College Environmental Studies Program

Title of the Study: Drinking Water and Septic Systems in the Messalonskee Lake Watershed

Researcher Name(s): Alyssa Kullberg (atkullbe@colby.edu) and Gail Carlson (gcarlson@colby.edu)

The general purpose of this research is get an idea as to the amount of people obtaining their drinking water from Messalonskee Lake, and how the risks may compare to other water sources, as well as describing septic systems in the watershed, as they are a potential source of pollutants. Participants in this study will be asked to answer a series of questions on the drinking water and septic system at their home.Informed consent is required by Colby College for any person participating in a College-sponsored research study. This study has been approved by the College's Institutional Review Board for Research with Human Subjects. I hereby give my consent to be the subject of this research study. I acknowledge that the researcher has provided me with:

A. An explanation of the study’s general purpose and procedure.
B. Answers to any questions I have asked about the study procedure.

I understand that:

A. My participation in this study will take approximately 5-10 minutes.
B. No unusual risks are anticipated as a result of participating in this research.
C. The potential benefits of this study include contributing to a greater understanding of water
quality issues in the Messalonskee Watershed.
D. I will not be compensated for participating in this study.
E. My participation is voluntary, and I may withdraw my consent and discontinue participation in the study at any time. My refusal to participate will not result in any penalty.

F. The specific nature of and reasons for the procedures employed, those aspects of my behavior that have been recorded for measurement purposes, and what the investigators hope to learn from this study will all be fully explained to me at the end of the session.
G. All data collected for this study will be kept confidential. The data will be stored in a secure location, and research reports will only present aggregate statistics without any personally identifying information.
H. I may, for any reason, choose to withhold use of any data provided by my participation.

Clicking on the AGREE button indicates that:
A. You have read the above information
B. You voluntarily agree to participate
C. You are 18 years of age or older

__AGREE
__DISAGREE

Is your residence in the Messalonskee Watershed seasonal or year-round?
- __ seasonal
- __ year-round

How do you obtain your drinking water?
- __ Directly from the lake
- __ Drilled well (100-400 ft)
- __ Driven well (30-50 ft)
- __ Dug well (10-30 ft)
- __ Bottled water
- __ Public water supply
- __ Don't know

If answered "Directly from the lake," directs to this question block:

Have you ever tested your water for contaminants?
- __ Yes
- __ No

When was the last time you tested your water? ______

Was the water sample tested straight from the lake or after filtration?
- __ From the lake
- __ After filtration

Were there high levels of any contaminants?
- __ Yes
- __ No

Which contaminants, and what was the concentration? _____

Do you filter or treat your water?
__ Yes
__ No

At what point do you filter or treat your water?
__ Point of entry into the home
__ For drinking only (at the tap or the sink)
__ Other (please specify) _____

Please describe your water filtration or treatment system. _____

If answered "Drilled well (100-400 ft)," "Driven well (30-50 ft)," or "Dug well(10-30 ft)," directs to this question block:

How old is your well? _____

Has your well ever been tested for arsenic?
__ Yes
__ No

When was the last time your well was tested for arsenic? _____

How much arsenic was detected in your well water? _____

Have you ever been told that you have too much arsenic in your well water?
__ Yes
__ No

Do you filter or treat your water?
__ Yes
__ No

At what point do you filter or treat your water?
__ Point of entry into the home
__ For drinking only (at the tap or the sink)
__ In the bathtub or shower
__ Other (please specify) ______

Please describe your water treatment or filtration system. _____

Do you have a septic system?
__ Yes
__ No

If you have more than one structure on your property, do you have one or multiple septic systems?

__ One septic system
__ Multiple
__ Not applicable

Do you know about how old your septic system is?

__ Yes
__ No

How old is your septic system?

__ 0 - 10 years old
__ 10 - 20 years old
__ > 20 years old

Does your septic system include a leach field / drain field?

__ Yes
__ No
__ Don't know

When is the last time your septic system was inspected? _____

Do you have your septic tank pumped regularly?

__ Yes
__ No

How frequently do you have your septic tank pumped? _____

When was the last time you had your septic tank pumped? _____

Have you ever heard about failing septic systems on Messalonskee Lake or septic seepage into Messalonskee Lake?

__ Yes
__ No

Please explain. _____

How serious of a problem do you think failing septic systems are on Messalonskee Lake?

__ Very serious
__ Somewhat serious
__ Not serious
__ Don't know

We would like to collect your street address for the sole purpose of making possible spatial analysis around the lake. Your address will not be made public, used for identification purposes, or listed in any publication or report.

What is your street address? _____

How long have you lived at your current residence? _____

If you would like to be provided with the results and conclusions of this study, please provide your email address (not required). _____

Thank you for taking the time to participate in this survey.

This information will be used in a study on Messalonskee Lake conducted by scientists from Colby College. We are primarily investigating the presence of certain pharmaceuticals and personal care products in the lake. Other than direct human input at high usage areas of the lake, it is possible that old and/or unmaintained septic systems in the watershed are contributing to pollutants that enter the lake. As there are few public records available on septic systems in the area, this information is invaluable not only to this study, but also for future studies and for better public knowledge of the state of septic systems in the area.

Depending on the presence and levels of pharmaceuticals and personal care products in the lake, it is possible that this issue also presents a public health concern, especially for those obtaining their drinking water directly from the lake. For this reason, we want an idea of how many people are obtaining their water this way, whether they are taking measures to sanitize their water, and how the risks might compare to those getting their water from a well. Friends of Messalonskee will likely distribute the findings of the study. If you have any questions relating to this study, do not hesitate to contact us.

Alyssa Kullberg (atkullbe@colby.edu)
Gail Carlson (gcarlson@colby.edu)

If you have any concerns about your rights as a participant in this study, please contact the Chair of the Colby Institutional Review Board for Research with Human Subjects, Tarja Raag (tarja.raag@colby.edu).

Thank you again for participating!

APPENDIX 3. Sampling site conditions by date, including surface (0.5 m) temperature and dissolved oxygen (DO) readings, basic weather descriptions, and other notes.

Date	Location	DO % saturation	DO concentration (mg/L)	temp (°C)	Weather	Notes
6/26/2017	ML1	102.1	8.6	23.7	sunny	
6/26/2017	ML2	104.3	8.8	23.8	sunny	
6/26/2017	ML3	106.0	8.9	23.9	sunny	
6/26/2017	ML4	108.7	9.3	23.3	sunny	
6/26/2017	ML5	107.0	9.3	22.5	sunny	
6/26/2017	ML6	106.1	9.2	22.4	sunny	
6/26/2017	ML7	109.7	9.3	23.8	sunny	
6/26/2017	ML8	109.6	9.4	23.1	sunny	
6/26/2017	ML9	104.9	9.1	22.5	sunny	
6/26/2017	ML10	103.6	9.0	22.4	sunny	
6/26/2017	ML11	33.9	2.9	23.7	sunny	
6/26/2017	ML12	95.1	8.0	23.9	sunny	
6/26/2017	ML13	110.2	9.5	22.9	sunny	
7/10/2017	ML6	102.0	8.6	23.7	partly cloudy, windy WSW ~12-15 mph *choppy	
7/10/2017	ML5	102.4	8.6	23.9	partly cloudy, windy WSW ~12-15 mph *choppy	
7/10/2017	ML4	103.7	8.6	25.1	partly cloudy, windy WSW ~12-15 mph *choppy	
7/10/2017	ML7	102.7	8.5	25.2	partly cloudy	Monday very quiet on the lake (saw 3 boats total) after a very busy weekend (Ned Hammond)
7/10/2017	ML8	104.6	8.7	24.7	partly cloudy	
7/10/2017	ML9	102.7	8.6	24.3	partly sunny	
7/10/2017	ML10	100.9	8.5	23.8	partly sunny	
7/10/2017	ML12	96.5	8.0	25.2	sunny	
7/10/2017	ML11	32.5	2.7	24.0	sunny	Several people smoking
7/10/2017	ML13	101.2	8.6	23.7	sunny *choppy	
7/10/2017	ML3	101.2	8.2	25.7	sunny	Flock of geese in water, lots of goose poop. Very litte buffer between Hammond Lumber and stream
7/10/2017	ML2	102.6	8.4	25.7	sunny	Water recently churned up by milfoil divers, extremely murky/opaque. Black tern breeding
7/10/2017	ML1	103.5	8.4	25.8	partly sunny	

7/24/2017	ML1	95.5	7.9	25.3	cloudy, cool, slightly breezy	A mom asked how many kids had peed in the lake, 3 kids raised hands
7/24/2017	ML2	97.5	8.0	25.4	cloudy, cool, slightly breezy	
7/24/2017	ML3	97.5	8.0	25.4	cloudy, cool, slightly breezy	Several people smoking, water clear.
7/24/2017	ML6	102.5	8.5	24.7	cloudy, cool, slightly breezy	
7/24/2017	ML5	101.8	8.5	24.6	cloudy, cool, slightly breezy	4 people fishing (kids), lots of fast-food trash, clear water
7/24/2017	ML4	100.1	8.2	25.3	cloudy, cool, slightly breezy	
7/24/2017	ML7	96.8	8.0	25.3	cloudy, cool, slightly breezy	
7/24/2017	ML8	101.5	8.4	25.2	cloudy, cool, slightly breezy	
7/24/2017	ML9	102.6	8.5	24.7	partly cloudy	morning after Oakfest triathlon (water portion in kayak); dog in water; porta-potties smelly "It happens every weekend" (Ned Hammond) that they overflow
7/24/2017	ML10	100.1	8.4	24.3	partly cloudy	(no gloves worn)
7/24/2017	ML11	29.3	2.5	22.8	partly cloudly	
7/24/2017	ML12	86.8	7.2	24.7	partly cloudy	MLRC crew sampling at buoy
7/24/2017	ML13	103.7	8.6	25.1	partly cloudy	
8/7/2017	ML1	95.4	7.9	24.7	partly sunny, breeze	MLRC crew had been there earlier
8/7/2017	ML2	97.3	8.0	25.0	partly sunny, breeze	
8/7/2017	ML3	99.1	8.1	25.3	partly sunny, breeze	
8/7/2017	ML6	97.1	8.2	23.8	partly sunny, breezy, wavy	
8/7/2017	ML5	98.4	8.3	24.0	partly sunny, breezy, wavy	
8/7/2017	ML4	100.2	8.3	25.0	partly sunny, breezy, wavy	
8/7/2017	ML7	100.1	8.3	25.1	partly sunny, breezy, wavy	
8/7/2017	ML8	101.1	8.4	24.5	partly sunny, breezy, wavy	
8/7/2017	ML9	100.2	8.4	24.0	partly sunny, breezy, wavy	
8/7/2017	ML10	96.8	8.3	23.0	partly sunny, breezy, wavy	
8/7/2017	ML11	28.8	2.5	22.3	calm, sunny	
8/7/2017	ML12	89.0	7.6	23.5	sunny	
8/7/2017	ML13	103.6	8.6	25.0	sunny, wavy	
8/9/2017	KR1	111.5	9.3	24.8	partly cloudy	

8/9/2017	KR2	109.6	9.0	25.2	partly cloudy
8/9/2017	MS2	79.8	6.8	23.5	partly cloudy
8/9/2017	MS1	89.7	7.4	25.0	partly cloudy

APPENDIX 4. A table of sample information, including each sample ID with site information and all measured concentrations of the 18 PPCPs involved in this study.

Sample ID	Site Location	Site Name	Site Use	Latitude	Longitude	Collection Date
ML1-1	ML1	Dam	PublicLow	44.540055	-69.723163	6/26/2017
ML2-1	ML2	Oakland swim area	PublicHigh	44.540209	-69.727102	6/26/2017
ML3-1	ML3	Oakland boat launch	PublicHigh	44.540504	-69.726241	6/26/2017
ML4-1	ML4	Hammond	OpenWater	44.5256	-69.744032	6/26/2017
ML5-1	ML5	MESSDEP2 (Oakland)	OpenWater	44.51951	-69.75059	6/26/2017
ML6-1	ML6	MESSDEP1 (Sidney)	OpenWater	44.49774	-69.77421	6/26/2017
ML7-1	ML7	Middle of lake	Private	44.487408	-69.786192	6/26/2017
ML8-1	ML8	Hume Center	PublicLow	44.510067	-69.759537	6/26/2017
ML9-1	ML9	Sidney boat launch	PublicHigh	44.490754	-69.77667	6/26/2017
ML10-1	ML10	106 Orchid Lane	Private	44.460372	-69.794611	6/26/2017
ML11-1	ML11	Belgrade boat launch	PublicHigh	44.44259	-69.83079	6/26/2017
ML12-1	ML12	Loon Ridge (stream)	Private	44.441866	-69.832243	6/26/2017
ML13-1	ML13	Townsend	Private	44.491503	-69.791529	6/26/2017
ML14-1	ML8	Hume Center	Private	44.510067	-69.759537	6/26/2017
ML15-1	ML9	Sidney boat launch	PublicHigh	44.44259	-69.83079	6/26/2017
ML16-1	ML12	Loon Ridge (stream)	Private	44.441866	-69.832243	6/26/2017
ML1-2	ML1	Dam	PublicLow	44.540055	-69.723163	7/10/2017
ML2-2	ML2	Oakland swim area	PublicHigh	44.540209	-69.727102	7/10/2017
ML3-2	ML3	Oakland boat launch	PublicHigh	44.540504	-69.726241	7/10/2017
ML4-2	ML4	Hammond	OpenWater	44.5256	-69.744032	7/10/2017
ML5-2	ML5	MESSDEP2 (Oakland)	OpenWater	44.51951	-69.75059	7/10/2017
ML6-2	ML6	MESSDEP1 (Sidney)	OpenWater	44.49774	-69.77421	7/10/2017
ML7-2	ML7	Middle of lake	Private	44.487408	-69.786192	7/10/2017
ML8-2	ML8	Hume Center	PublicLow	44.510067	-69.759537	7/10/2017
ML9-2	ML9	Sidney boat launch	PublicHigh	44.490754	-69.77667	7/10/2017
ML10-2	ML10	106 Orchid Lane	Private	44.460372	-69.794611	7/10/2017
ML11-2	ML11	Belgrade boat launch	PublicHigh	44.44259	-69.83079	7/10/2017

ML12-2	ML12	Loon Ridge (stream)	Private	44.441866	-69.832243	7/10/2017
ML13-2	ML13	Townsend	Private	44.491503	-69.791529	7/10/2017
ML14-2	ML3	Oakland boat launch	PublicHigh	44.540504	-69.726241	7/10/2017
ML15-2	ML2	Oakland swim area	PublicHigh	44.540209	-69.727102	7/10/2017
ML16-2	ML1	Dam	PublicLow	44.540055	-69.723163	7/10/2017
ML1-3	ML1	Dam	PublicLow	44.540055	-69.723163	7/24/2017
ML2-3	ML2	Oakland swim area	PublicHigh	44.540209	-69.727102	7/24/2017
ML3-3	ML3	Oakland boat launch	PublicHigh	44.540504	-69.726241	7/24/2017
ML4-3	ML4	Hammond	OpenWater	44.5256	-69.744032	7/24/2017
ML5-3	ML5	MESSDEP2 (Oakland)	OpenWater	44.51951	-69.75059	7/24/2017
ML6-3	ML6	MESSDEP1 (Sidney)	OpenWater	44.49774	-69.77421	7/24/2017
ML7-3	ML7	Middle of lake	Private	44.487408	-69.786192	7/24/2017
ML8-3	ML8	Hume Center	PublicLow	44.510067	-69.759537	7/24/2017
ML9-3	ML9	Sidney boat launch	PublicHigh	44.490754	-69.77667	7/24/2017
ML10-3	ML10	106 Orchid Lane	Private	44.460372	-69.794611	7/24/2017
ML11-3	ML11	Belgrade boat launch	PublicHigh	44.44259	-69.83079	7/24/2017
ML12-3	ML12	Loon Ridge (stream)	Private	44.441866	-69.832243	7/24/2017
ML13-3	ML13	Townsend	Private	44.491503	-69.791529	7/24/2017
ML14-3	ML6	MESSDEP1 (Sidney)	OpenWater	44.49774	-69.77421	7/24/2017
ML15-3	ML5	MESSDEP2 (Oakland)	OpenWater	44.51951	-69.75059	7/24/2017
ML16-3	ML4	Hammond	OpenWater	44.5256	-69.744032	7/24/2017
ML17-3	ML7	Middle of lake	Private	44.487408	-69.786192	7/24/2017
ML1-4	ML1	Dam	PublicLow	44.540055	-69.723163	8/7/2017
ML2-4	ML2	Oakland swim area	PublicHigh	44.540209	-69.727102	8/7/2017
ML3-4	ML3	Oakland boat launch	PublicHigh	44.540504	-69.726241	8/7/2017
ML4-4	ML4	Hammond	OpenWater	44.5256	-69.744032	8/7/2017
ML5-4	ML5	MESSDEP2 (Oakland)	OpenWater	44.51951	-69.75059	8/7/2017
ML6-4	ML6	MESSDEP1 (Sidney)	OpenWater	44.49774	-69.77421	8/7/2017
ML7-4	ML7	Middle of lake	Private	44.487408	-69.786192	8/7/2017
ML8-4	ML8	Hume Center	PublicLow	44.510067	-69.759537	8/7/2017
ML9-4	ML9	Sidney boat launch	PublicHigh	44.490754	-69.77667	8/7/2017

ML10-4	ML10	106 Orchid Lane	Private	44.460372	-69.794611	8/7/2017
ML11-4	ML11	Belgrade boat launch	PublicHigh	44.44259	-69.83079	8/7/2017
ML12-4	ML12	Loon Ridge (stream)	Private	44.441866	-69.832243	8/7/2017
ML13-4	ML13	Townsend	Private	44.491503	-69.791529	8/7/2017
ML14-4	ML11	Belgrade boat launch	PublicHigh	44.490754	-69.77667	8/7/2017
ML15-4	ML10	106 Orchid Lane	Private	44.460372	-69.794611	8/7/2017
ML16-4	ML13	Townsend	Private	44.491503	-69.791529	8/7/2017
KR1	KR1	Downstream	--	44.524461	-69.656632	8/9/2017
KR3	KR1	Downstream	--	44.524461	-69.656632	8/9/2017
KR2	KR2	Upstream	--	44.533175	-69.644094	8/9/2017
KR4	KR2	Upstream	--	44.533175	-69.644094	8/9/2017
MS1	MS1	North St. boat launch	--	44.564622	-69.648747	8/9/2017
MS2	MS1	North St. boat launch	--	44.564622	-69.648747	8/9/2017

APPENDIX 4 (continued). A table of sample information, including each sample ID with site information and all measured concentrations of the 18 PPCPs involved in this study. All concentrations are in µg/L.

Sample ID	1,7-Dimethylxanthine	Acetaminophen	Amphetamine	Caffeine	Carbamazepine	Cimetidine	Cotinine	Diphenhydramine	MDA	MDMA	Methamphetamine	Morphine	Phenazone	Sulfachloropyridazine	Sulfamethazine	Sulfamethoxazole	Thiabendazole	Trimethoprim	% Orphenadrine
ML1-1	0.001	0.001	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.005	0.000	0.000	0.000	0.000	61.7
ML2-1	0.001	0.001	0.004	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.007	0.000	0.000	0.000	0.000	86.2
ML3-1	0.001	0.001	0.006	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.005	0.000	0.000	0.000	0.000	120.1
ML4-1	0.001	0.001	0.012	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.006	0.000	0.000	0.000	0.000	105.1
ML5-1	0.001	0.000	0.025	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.005	0.000	0.000	0.000	0.000	73.9
ML6-1	0.001	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.007	0.000	0.000	0.000	0.000	89.1
ML7-1	0.001	0.000	0.004	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.000	0.000	0.000	0.000	104.2
ML8-1	0.000	0.000	0.003	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	102.2
ML9-1	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	99.5
ML10-1	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	72.3
ML11-1	0.001	0.002	0.061	0.007	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.000	0.000	0.000	0.000	115.1
ML12-1	0.001	0.000	0.002	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	118.0
ML13-1	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	89.6
ML14-1	0.000	0.000	0.024	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	58.3
ML15-1	0.001	0.001	0.010	0.005	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	139.2
ML16-1	0.001	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	118.7
ML1-2	0.001	0.002	0.001	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	117.0
ML2-2	0.001	0.002	0.002	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	71.7
ML3-2	0.023	0.008	0.001	0.020	0.000	0.000	0.003	0.003	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	115.5
ML4-2	0.001	0.001	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	110.5
ML5-2	0.001	0.000	0.002	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	101.7
ML6-2	0.000	0.000	0.002	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	95.8
ML7-2	0.001	0.002	0.001	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	99.1

ML8-2	0.001	0.000	0.004	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	89.8
ML9-2	0.001	0.000	0.016	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	101.2
ML10-2	0.000	0.000	0.022	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	114.0
ML11-2	0.004	0.000	0.011	0.009	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	93.1
ML12-2	0.001	0.000	0.006	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	124.5
ML13-2	0.000	0.000	0.002	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	57.2
ML14-2	0.018	0.006	0.002	0.017	0.000	0.000	0.002	0.002	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	103.4
ML15-2	0.001	0.002	0.002	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	101.3
ML16-2	0.001	0.002	0.004	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	98.1
ML1-3	0.001	0.003	0.004	0.004	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	97.6
ML2-3	0.001	0.005	0.001	0.003	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	90.6
ML3-3	0.002	0.007	0.007	0.003	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	81.5
ML4-3	0.001	0.000	0.002	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	110.6
ML5-3	0.001	0.000	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	86.4
ML6-3	0.001	0.000	0.002	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	92.0
ML7-3	0.001	0.001	0.001	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	90.5
ML8-3	0.001	0.000	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	93.1
ML9-3	0.001	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	111.3
ML10-3	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	123.7
ML11-3	0.003	0.001	0.084	0.007	0.000	0.000	0.004	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	217.3
ML12-3	0.000	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	144.5
ML13-3	0.001	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	91.8
ML14-3	0.001	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	101.9
ML15-3	0.000	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	87.5
ML16-3	0.001	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	98.2
ML17-3	0.001	0.001	0.003	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	94.7
ML1-4	0.001	0.001	0.001	0.002	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	98.7
ML2-4	0.001	0.001	0.002	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	108.2
ML3-4	0.001	0.001	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	94.9
ML4-4	0.001	0.000	0.003	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	77.8
ML5-4	0.000	0.000	0.003	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	102.5

ML6-4	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	104.7
ML7-4	0.001	0.001	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	82.5
ML8-4	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	96.3
ML9-4	0.000	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	93.8
ML10-4	0.001	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	41.2
ML11-4	0.002	0.000	0.236	0.003	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	33.4
ML12-4	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	110.9
ML13-4	0.001	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	81.2
ML14-4	0.000	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	92.2
ML15-4	0.001	0.000	0.000	0.001	0.000	0.000	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	88.1
ML16-4	0.001	0.000	0.000	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	120.8
KR1	0.002	0.010	0.000	0.017	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	98.1
KR3	0.003	0.012	0.000	0.018	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	85.8
KR2	0.001	0.001	0.000	0.008	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	111.7
KR4	0.001	0.001	0.000	0.008	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	91.9
MS1	0.001	0.003	0.000	0.035	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	97.8
MS2	0.001	0.003	0.000	0.034	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	119.5

APPENDIX 5. Total nitrogen (TN) and total phosphorus (TP) concentration measurements per sample, listed by site. Samples for TN and TP measurements were taken on 7/24/2017 and 8/7/2017. Outlier data points have been omitted.

Date	Sample Location	TN Sample Avg. (mg/L)	TN Std. Err.	TP Sample Avg. (mg/L)	TP Std. Err.
7/24/2017	ML1	0.295	0.0060	0.023	0.0024
7/24/2017	ML1	0.451	0.0250	0.020	0.0032
8/7/2017	ML1	0.321	0.0020	0.026	0.0006
8/7/2017	ML1	0.436	0.0045	0.025	0.0008
7/24/2017	ML2	0.424	0.0020	--	--
7/24/2017	ML2	0.378	0.0015	0.015	0.0002
8/7/2017	ML2	0.373	0.0085	0.017	0.0012
8/7/2017	ML2	0.224	0.0135	0.021	0.0001
7/24/2017	ML3	0.400	0.0035	0.028	0.0033
7/24/2017	ML3	0.514	0.0360	0.018	0.0010
8/7/2017	ML3	0.362	0.0130	0.020	0.0004
8/7/2017	ML3	0.328	0.0135	0.018	0.0002
7/24/2017	ML4	0.399	0.0330	0.022	0.0039
7/24/2017	ML4	0.410	0.0010	0.023	0.0010
8/7/2017	ML4	0.320	0.0310	0.025	0.0011
8/7/2017	ML4	0.373	0.0000	0.014	0.0009
7/24/2017	ML5	0.404	0.0010	0.016	0.0015
7/24/2017	ML5	0.435	0.0585	0.020	0.0013
8/7/2017	ML5	--	--	0.024	0.0005
8/7/2017	ML5	0.345	0.0010	0.024	0.0067
7/24/2017	ML6	0.373	0.0185	0.016	0.0001
7/24/2017	ML6	0.366	0.0475	--	--
8/7/2017	ML6	0.381	0.0085	0.015	0.0024
8/7/2017	ML6	0.230	0.0120	0.025	0.0010
7/24/2017	ML7	0.219	0.0425	0.024	0.0001
7/24/2017	ML7	0.349	0.0140	--	--
8/7/2017	ML7	0.420	0.0035	0.013	0.0005
8/7/2017	ML7	0.381	0.0035	0.022	0.0019
7/24/2017	ML8	0.272	0.0400	0.016	0.0002
7/24/2017	ML8	0.402	0.0040	0.022	0.0000
8/7/2017	ML8	0.357	0.0000	0.019	0.0001
8/7/2017	ML8	0.388	0.0100	0.019	0.0055
7/24/2017	ML9	0.240	0.0120	0.024	0.0009
7/24/2017	ML9	0.375	0.0010	0.016	0.0024
8/7/2017	ML9	0.535	0.0425	0.031	0.0020
8/7/2017	ML9	0.346	0.0135	0.022	0.0002
7/24/2017	ML10	0.385	0.0210	0.015	0.0000
7/24/2017	ML10	0.392	0.0110	0.016	0.0003
8/7/2017	ML10	0.249	0.0130	0.018	0.0016

8/7/2017	ML10	0.198	0.0185	0.024	0.0033
7/24/2017	ML11	0.515	0.0020	0.027	0.0018
7/24/2017	ML11	0.514	0.0020	0.023	0.0052
8/7/2017	ML11	0.423	0.0010	0.032	0.0059
8/7/2017	ML11	0.477	0.0110	0.032	0.0071
7/24/2017	ML12	0.420	0.0015	0.023	0.0000
7/24/2017	ML12	0.445	0.0300	0.029	0.0021
8/7/2017	ML12	0.373	0.0025	0.026	0.0011
8/7/2017	ML12	0.367	0.0330	0.020	0.0001
7/24/2017	ML13	0.419	0.0010	0.022	0.0029
7/24/2017	ML13	0.331	0.0055	0.020	--
8/7/2017	ML13	0.363	0.0080	0.018	0.0002
8/7/2017	ML13	0.402	0.0040	0.021	0.0018

APPENDIX 6. Respondent data from the septic system and drinking water survey conducted among Messalonskee Lake Watershed residents. To see the original wording and format of each question, see Appendix 1.

Respondent	Year-round residence?	How long lived on/owned property?	Drinking water source	Septic system?	One or multiple sys.?	Do you know the septic sys.. age?	How old is your septic sys.?	Septic sys. leach field?	Time since septic inspection (years)
1	--	40	1	1	--	1	2	--	13
2	1	10	3	1	1	1	0	1	0
3	1	16	3	1	1	1	2	1	2
4	1	57	3	1	0	1	2	1	32
5	1	17	3	1	1	1	1	1	2
6	0	8	3	1	1	0	NA	1	10
7	1	15	3	1	1	1	2	1	10
8	0	15	3	1	0	1	2	1	1
9	1	16	3	1	--	1	1	1	1
10	1	17	3	1	1	1	1	1	0
11	0	16	1	1	--	0	NA	1	5
12	0	4	1	1	1	1	1	1	5
13	0	60	3	1	1	1	1	1	2
14	0	31	3	1	--	1	2	1	3
15	0	14	1	1	--	1	2	1	1
16	1	50	3	1	1	1	0	1	5
17	0	10	1	1	--	1	1	1	3
18	1	20	0	1	--	1	0	1	6
19	1	10	1	1	0	1	0	0	4
20	0	40	2	0	NA	NA	NA	NA	NA
21	1	12	3	1	--	1	2	1	12
22	1	2	3	1	1	1	0	1	2
23	0	1	1	1	1	1	0	1	1
24	1	46	1	1	--	1	0	1	1
25	1	28	3	1	1	0	NA	1	25

26	0	21	4	1	1	1	1	1	2
27	1	1	3	1	1	1	0	1	--
28	0	52	1	1	1	1	2	1	--
29	1	25	3	1	0	1	0	1	--
30	0	--	5	1	--	1	2	1	--
31	1	19	3	1	--	1	1	1	19
32	1	4	3	1	1	1	0	1	2
33	1	3	3	1	--	1	1	1	0
	0 = seasonal 1 = yr-round		0 = don’t know 1 = bottled 2 = dug well 3 = drilled well 4 = lake	0 = no 1 = yes	0 = >1 1 = one	0 = no 1 = yes	0 = <10 years 1 = 10-20 years 2 = >20 years		

APPENDIX 6 (continued). Respondent data from the septic system and drinking water survey conducted among Messalonskee Lake Watershed residents. To see the original wording and format of each question, see Appendix 1.

Res. #	Septic sys. pumped regularly?	Septic sys. pump frequency (years)	Time since septic sys. pumped (years)	Heard of septic sys. failure on lake?	If so, explain. (unedited responses)	Severity of septic sys. failure on lake?
1	1	2	2	0	NA	2
2	1	5	0	1	As Messalonskee's LakeSmart spokesperson, I check home owner's septic situation and have heard about past septic problems	3
3	1	2	1	0	NA	3
4	1	3	3	0	NA	0
5	1	2	2	1	Previous house we owned had a failed system and prior to occupancy we had a whole new system installed	0
6	1	3	3	0	NA	1
7	1	3	3	0	NA	1
8	0	7.5	2	0	NA	0
9	1	2	1	1	Ive lived on the lake for 40 years (my entire life) and have seen first hand septics seeping into the lake.	2
10	0	2	--	1	Check each side of me , one connected into my leach field , why i had septic guy come down, other has a barrel in the ground beside a seasonal stream!!barrel	3
11	1	3	0	0	NA	3
12	1	2	2	0	NA	3
13	1	3.5	2	1	They have been redone in last couple of years	3
14	1	5	3	0	NA	3
15	1	1	0	0	NA	0
16	1	3	1	1	Neighbors commenting on a summer residence. I'm not sure if I believe it.	2
17	1	2.5	2	0	NA	2

18	0	7	--	0	NA	0
19	1	4	--	0	NA	0
20	NA	NA	NA	0	NA	2
21	1	4	3	1	Just news from lake associations.	2
22	1	3	2	1	word of mouth only	3
23	0	4	--	0	NA	0
24	1	1	0	0	NA	0
25	0	25	--	1	My landlord says, as long as you don't have a problem, don't bother.	0
26	1	2	3	0	NA	3
27	--	--	--	1	I'm in real estate	2
28	0	--	--	0	NA	0
29	1	2.5	0	0	NA	3
30	0	--	--	0	NA	0
31	1	1	1	0	NA	2
32	1	4	2	1	My neigbors system failed last year, they had to dig up a section and have it pumped. Not sure if anything got in the lake.	2
33	1	2	0	0	NA	0
						0 = don’t know 1 = not serious 2 = somewhat serious 3 = very serious