Occurrence of Nitrate and Indicators of Agricultural and Septic System Contamination in a West Central Wisconsin Sand Aquifer

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Abstract **Fertilizers, manure, and septic effluent are potential sources of nitrate in groundwater. Nitrate can be harmful if ingested above the U.S. Environmental Protection Agency maximum contaminant level of 10 mg/L. In Eau Claire County, located in West Central Wisconsin, approximately one quarter of households rely on private wells. Sources of nitrate in private wells in Eau Claire County have not been researched previously.**

A total of 110 private wells in Eau Claire County were tested for seven agricultural and three septic indicators to identify sources of nitrate contamination. Nitrate contamination risk factor data (e.g., well depth, casing depth) were also collected. Average nitrate concentrations were significantly higher in wells with agricultural indicators, suggesting agriculture is a source of nitrate. Wastewater indicators were identified, but septic systems were not a significant source of nitrate. Well casing depth was the only risk factor associated with elevated nitrate. Funds should be allocated to the Eau Claire City–County Health Department to promote and subsidize point-of-use drinking water treatment in homes with nitrate levels ≥10 mg/L. Further, new well casing depths should be >12 m (40 ft) to avoid infiltration of nitrate and other contaminants.

Introduction

Nitrate is a widespread, highly mobile contaminant of groundwater that is especially common in dense agricultural areas (Spalding & Exner, 1993). Potential sources of nitrate contamination include agricultural or lawn fertilizer application, septic systems, animal feedlots and barnyards, and septage or sludge disposal. The burden of nitrate contamination in groundwater in the Upper Midwest has been widely studied (Bundy et al., 1996; Chern et al., 1999; LeMasters & Baldock, 1997; Shaw, 1994), partly because of the human health effects associated with nitrate exposure. Though nitrate is a naturally occurring compound, it is often found in groundwater at levels that greatly exceed the U.S. Environmental Protection Agency (U.S. EPA) preventive action limit (2 mg/L) or maximum contaminant level (MCL, 10 mg/L) in agricultural and dense unsewered residential areas.

The health-based standards for nitrate were established from the risk of methemoglobinemia, a condition in which the blood's ability to transport oxygen is compromised. Individuals who are pregnant and infants are at greatest risk. Some studies also suggest livestock that drink water with elevated nitrate have poorer pregnancy outcomes (Al-Qudah et al., 2009).

The Wisconsin Department of Natural Resources (WI DNR) has estimated that 90% of nitrate in Wisconsin groundwater is from agricultural activities, approximately 9% is from septic systems, and <1% is attributable to lawn fertilizer or other sources (Shaw, 1994). In addition to the health risks from nitrate, there could be additional risks to private well owners where co-contaminants associated with agriculture and septic systems exceed preventative action limits. Elevated nitrate often is correlated with pesticides, herbicides, viruses, pharmaceuticals, or other constituents of agrichemicals or human wastewater (Burow et al., 1998; Istok et al., 1993; Seiler et al., 1999). One study estimated that 42% of private drinking water wells in Wisconsin contained a detectable level of an herbicide or herbicide metabolite (Wisconsin Department of Agriculture, Trade, and Consumer Protection [WI DATCP], 2017).

In Eau Claire County located in West Central Wisconsin, over 25,000 people (approximately 1 in 4) rely on private wells as their primary source of drinking water. The quality of private well water is of public health concern because private water supplies are not regularly tested or regulated. Over 4,500 nitrate tests have been analyzed at the Eau Claire City–County Health Department (ECC–CHD) since 2005. Approximately 4,500 wells remain untested in Eau Claire County for nitrate. Approximately

1 in 2 wells sampled in Eau Claire County have nitrate that exceeds naturally occurring concentrations (generally presumed to be ≤2 mg/L). Nearly 1 in 20 sampled wells exceed the health-based standard for nitrate.

Until our study, almost no wells had been tested for common nitrate co-contaminants such as pharmaceuticals or agricultural chemicals in Eau Claire County, though other areas of the state have been investigated as early as the 1980s (Rothschild et al., 1982). Aims of our study were to determine nitrate trends and identify nitrate contamination risk factors of private wells in Eau Claire County.

Methods

Site Selection

This study took place from July 2016 through June 2018 and was approved by the University of Wisconsin–Eau Claire Institutional Review Board in 2016. Private well owners with a septic system and past water test containing nitrate levels ≥5 mg/L in the ECC–CHD Certified Public Health Laboratory water quality database were invited to participate. This level was used because it provided a robust number of potential participants (399) and at ≥5 mg/L, the nitrate present was likely from an anthropogenic source (U.S. Geological Survey, 1999). Well owners were mailed a letter describing the study that contained instructions to contact ECC–CHD to participate in the study.

Questionnaire

We developed the questionnaire used in our study in consultation with researchers from a similar study in Hastings, Minnesota, to create an exhaustive list of potential risk factors of nitrate contamination of well water (Dakota County Environmental Management, 2003). Property owners were issued a questionnaire on-site that gathered well contamination risk factor data such as site history and proximity to potential nitrate sources (e.g., septic systems, distance to and type of agricultural fields, fertilizer storage, abandoned wells). Using the questionnaire, researchers also recorded approximate distances from the wellhead to potential sources of nitrate such as animal feedlots, privies, and fertilizer storage. For each well, we gathered construction data—including well depth, construction date, casing depth, and well type—prior to sampling wells that had records available from WI DNR.

Sample Collection and Analysis

We collected samples for nitrate as well as seven agricultural indicators (i.e., atrazine, desethyl atrazine, desisopropyl atrazine, acetochlor, alachlor, metolachlor, and cyanazine) and three septic system indicators (i.e., caffeine, carbamazepine, and carisoprodol). We collected water samples for nitrate in clear, sterilized 250-ml polyethylene bottles. We collected agricultural and septic system indicator samples in 1-L amber glass bottles.

Samples were collected from an outside tap or pressure tank tap (before in-line water treatment systems where present) and after running the source for approximately 2 min. If no water treatment system was present, we also collected water samples from the indoor tap. Samples were transported on ice to the ECC–CHD laboratory, stored at <6 °C, and processed within 24 hr of collection.

Nitrate samples were analyzed using Standard Method 4500D-NO_3. Nitrate standards were prepared from pure potassium nitrate (Fisher Scientific). Nitrate standards and samples were treated with an interference suppressor and then analyzed with a calibrated ion-selective electrode.

Target chemicals for agricultural and septic system indicators were obtained as neat standards and prepared as diluted solutions in ethyl acetate (ChemService). Samples were analyzed using modified U.S. EPA (1995) Method 507. Control spikes were prepared by addition of standard solutions to 1 L of reagent water. Method blanks consisted of 1 L reagent water. To aid in recovery, 50 g of sodium chloride was dissolved in the samples, and 1,2-dimethyl-3-nitrobenzene was added as a surrogate spike. A sample size of 1 L was drawn through a Empore C18 and an SPD-RPD extraction disk (3M). The disks were eluted first with 8 ml ethyl acetate and then with 8 ml methylene chloride. The eluant was dried with sodium sulfate powder then reduced to 5 ml volume by evaporation of the solvent over a hot plate at 100 °C until the volume was reduced to 5 ml. The extract was injected into a calibrated Trace 1300 gas chromatograph with a nitrogen–phosphorus detector to determine the sample concentration (Thermo Fisher Scientific). Both the control spikes and method blanks (one of each per batch) were processed in the same manner as the samples. Hydrocodone, acetaminophen, flumetsulam, mesotrione, saccharin, and sulfamethoxazole were evaluated as potential indicator compounds—but were not amenable to the method.

Statistical Analysis

A student's *t*-test at the 95% confidence level was performed on dichotomous questionnaire responses to determine if the average nitrate concentration differed among sites with agricultural or septic system indicators and risk factors identified on the questionnaire. For example, the average nitrate concentration was compared at sites positive and negative for agricultural indicators to determine if herbicides and pesticides are indicators of nitrate contamination in private wells. Pearson's correlation coefficient was used to explore associations between numerical data collected on the questionnaire. Correlation coefficients (r) > .3 and < .5 indicate a moderate correlation and r > .5 indicates a strong correlation. STATA data analysis and statistical software version 13.1 was used to perform the statistical analysis.

Results

Sample Demographics

There were 399 eligible participants for our study. Of these, 130 households indicated interest (33% response rate) and 110 fully participated by completing the questionnaire and submitting water samples (28% response rate). A total of 108 samples were above the nitrate, agricultural, or septic system indicator detection limits; thus, we included these 108 samples in statistical analysis. Samples were collected from 10 different townships in Eau Claire County, with an additional 3 county townships having no participants. Positive samples of agricultural and septic systems were limited to 3 townships (Table 1). No agricultural or human waste indicators were found in samples from the other 7 townships.

Agricultural and Septic System Indicators

Agricultural indicators were identified in 15% of samples; septic system indicators were found in 5% of samples. Agricultural indicators detected were desethyl atrazine, desisopropyl atrazine, atrazine, and alachlor. Detected septic system indicators included caffeine and carbamazepine (Table 2). The most frequent agricultural indicator was desethyl atrazine (13% of samples), followed by atrazine (10% of samples). Of the 108 samples, 16 samples (15%) were positive for atrazine and/or an atrazine metabolite and 1 sample was positive for alachlor. Caffeine was the most frequent septic system indicator (4%). The four sites with caffeine detections were independent from the two sites with carbamazepine detects (the only other detected human waste indicator). Of the sites with atrazine detects, only two did not have atrazine metabolite detects. The four sites with atrazine metabolite detects did not have atrazine present in groundwater at detectable levels.

Nitrate

The nitrate MCL was exceeded in 24 of 108 samples (22%). The maximum detected nitrate concentration was more than double the MCL at 22 mg/L. The average nitrate concentrations in each township are shown in Figure 1. None of the agricultural or septic system indicators was above available enforcement standards. The average nitrate concentration in wells with agricultural indicators present was 10.7 mg/L, which is significantly higher at the 95% confidence level than the average nitrate concentration in wells without agricultural indicators present (6.8 mg/L; $p < .0026$).

The median nitrate concentration was 6.7 mg/L. When comparing the average nitrate concentration in wells positive for atrazine (but no other agricultural indicators) with wells without atrazine, nitrate concentrations were significantly higher in atrazine wells ($p < .0025$). No statistically significant relationship was found between wells with high nitrate concentrations and presence of the septic system indicators analyzed.

Nitrate Contamination Risk Factors

Contrary to our hypothesis, there were weak correlations between nitrate concentration and well age ($r = .08$) and well depth ($r = .17$). Other analyzed variables with weak correlations to nitrate concentration were drillhole depth ($r = .21$), static water level ($r = .22$), and well screen length ($r = .05$). Well construction information was available for 39% of sampled sites. Among these sites, wells with a casing depth <12 m (40 ft) had significantly more nitrate at the 95% confidence level ($p < .032$). A total of 73% of households (52 households) that reported a crop within 91 m (300 ft) of the well stated the crop was corn.

TABLE **1**

Number of Samples Collected and Percentage of Samples Positive for Agricultural and Septic System Indicators, Eau Claire County Townships, Wisconsin

Township	Sample Size	Population	# of Permitted Septic and Holding Tanks	Samples Positive for Agricultural Indicators # (%)	Samples Positive for Septic System Indicators # (%)
Bridge Creek	3	1,902	699	0	0
Brunswick	16	1,713	700	0	0
Clear Creek	1	817	331	0	0
Drammen	2	745	330	0	0
Lincoln	3	1,186	444	0	0
Ludington	1	1,096	479	0	0
Pleasant Valley	27	3,181	1,355	6 (22)	3 (11)
Seymour	8	3,276	1,299	0	0
Union	14	2,736	1,071	10 (71)	1 (7)
Washington	29	7,379	2,278	1 (3)	2 (7)

Note. Population numbers were calculated from 2013–2017 U.S. Census Bureau estimates.

Discussion

Agricultural and Septic System Indicators

Atrazine and desethyl atrazine (an atrazine metabolite) were the most frequent agricultural indicators detected. The frequency of detection was similar to Wisconsin's state average. Throughout Wisconsin, atrazine and atrazine metabolites are present in approximately 23% of private wells compared with 15% in our study (WI DATCP, 2017). The infrequent detection of the other agriculture and septic system indicators could be due to a variety of reasons. Atrazine is a broadleaf herbicide for agriculture, and weed control is responsible for the overwhelming majority of atrazine in the environment. Because atrazine is classified as a restricted-use pesticide, only certified applicators are permitted to purchase or apply it. Atrazine is not very persistent in surface soils after application due to biodegradation. The half-life of atrazine in soil has been reported within a range of 14–109 days. Slow or no biodegradation occurs once atrazine is in groundwater (Agency for Toxic Substances and Disease Registry, 2011). The low number of atrazine detects in groundwater for our study is likely a result of its biodegradation prior to entering the water column.

Caffeine and carbamazepine were the only septic system indicators detected. Caffeine can serve as an effective indicator of groundwater contamination from septic systems because of its widespread use (Seiler et al., 1999). Caffeine might be present in wastewater as unmetabolized caffeine consumed in beverages or via disposal of unconsumed coffee, soft drinks, or tea. Of the sampled wells, four contained caffeine at detectable levels; the maximum concentration of caffeine was 0.36 µg/L, which is slightly higher than other similar studies. For example, Seiler et al. (1999) detected 0.23 µg/L of caffeine below an unsewered Nevada subdivision.

Considering 100% of sampled sites in our study have septic systems, a higher number of detectable concentrations of caffeine or other wastewater indicators was expected. Caffeine

TABLE **2**

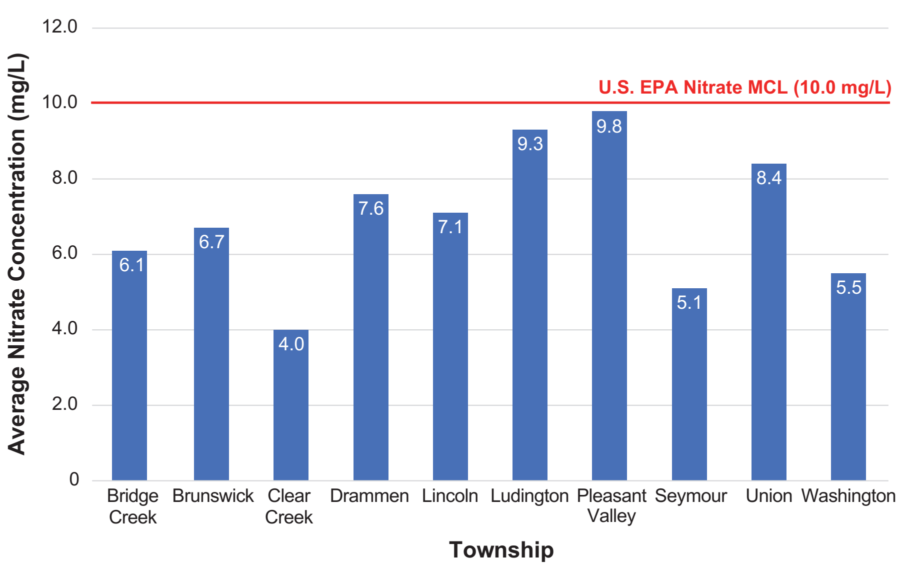
Maximum Concentrations, Frequency, and Detection Limits for the Analysis of Agricultural and Septic System Indicators and Nitrate, Eau Claire County, Wisconsin

Indicator	Chemical	Chemical Purpose	Detection Limit (µg/L)	Maximum Concentration Detected (µg/L)	MCL (µg/L)	# of Detects
Agricultural	Desethyl atrazine	Atrazine metabolite	0.2	0.49	NA	14
	Desisopropyl atrazine	Atrazine metabolite	0.2	0.42	NA	3
	Atrazine	Herbicide	0.1	0.49	3	11
	Acetochlor	Herbicide	0.2	ND	NA	0
	Alachlor	Herbicide	0.2	0.28	2	1
	Metolachlor	Herbicide	0.2	ND	NA	0
	Cyanazine	Herbicide	0.1	ND	NA	0
Septic system	Caffeine	Stimulant	0.2	0.36	NA	4
	Carbamazepine	Anticonvulsant	0.3	0.85	NA	2
	Carisoprodol	Muscle relaxant	0.3	ND	NA	0
	Nitrate	Fertilizer, waste product	0.41 mg/L	22 mg/L	10 mg/L	108

Note. MCL = maximum contaminant level; NA = not applicable; ND = not detected.

FIGURE **1**

Average Nitrate Concentrations in Private Wells, Eau Claire County Townships, Wisconsin



Note. No township's average nitrate concentration in private wells exceeded the U.S. Environmental Protection Agency (U.S. EPA) maximum contaminant level (MCL) for nitrate of 10 mg/L.

is highly biodegradable in soils with strong microbiological communities, however, and is known to sorb to sandy loam and silt loam soils, which are present in Eau Claire County (Karnjanapiboonwong et al., 2010; Knee et al., 2010) and might reduce the presence of caffeine in groundwater.

Conversely, carbamazepine does not degrade or sorb and can survive intact in groundwater for >8 years (Clara et al., 2004; Drewes et al., 2003). These properties make carbamazepine a good option for a wastewater tracer. Carbamazepine, however, is much less ubiquitous in septic systems compared to caffeine due to its overall lower rate of consumption. Seiler et al. (1999) found 1 positive sample for carbamazepine in 16 samples from unsewered subdivisions. Out of 38 groundwater sampling locations in western Montana, 11 locations contained detectable carbamazepine with a maximum concentration of 0.42 µg/L (Miller & Meek, 2006) in comparison with a maximum 0.85 µg/L found in our study.

The relatively low detection rates of caffeine and pharmaceuticals do not confirm that a well has not been impacted by septic effluent, though, especially given the transport barriers of caffeine and potential sporadic use of the pharmaceuticals. In future studies, better indicators of septic system impacts might be a) other chemicals that are less biodegradable and ubiquitous or b) pharmaceutical metabolites.

Nitrate

Across Wisconsin as a whole, 10–11% of private wells on average are above the nitrate MCL (LeMasters & Baldock, 1997; WI DATCP, 2017). Even though the sample population in our study contained wells known to have at least ≥5 mg/L nitrate, the percentage of samples above the U.S. EPA nitrate MCL for drinking water (10 mg/L) that we found (22%) is similar to what other regional studies found. In areas with abundant agriculture in Wisconsin, much like Eau Claire County, 17–26% of private wells contain nitrate above the U.S. EPA MCL (LeMasters & Baldock, 1997). In nearby Hastings, Minnesota, researchers found 25% of private and public drinking water wells had nitrate concentrations above the U.S. EPA MCL (Hastings is 140 km west of Eau Claire County) and deemed these findings as a water quality "problem" for the area (Dakota County Environmental Management, 2003).

For existing private wells in Eau Claire County with nitrate tests >10 mg/L, homeowners are notified and point-of-use or whole-house system installation is recommended by ECC–CHD. Nitrate testing of private wells in Eau Claire County, however, is not required, and there is no funding to help homeowners purchase point-of-use treatment systems.

Results from a statewide study found that 70% of Wisconsin homeowners did not take action to reduce nitrate drinking water exposures (Knobeloch et al., 1997). Among the homeowners who did take action in our study population, the most common solutions were purchasing bottled water and installing a point-of-use nitrate treatment system. The average cost of purchasing bottled water or installing a point-of-use treatment system at the time of the Knobeloch et al. (1997) study was $200/year and $850/year, respectively.

Present-day estimates for bottled water (1 gallon/day) are approximately $475/person/year. Reverse osmosis systems are available currently for a one-time cost of at least $200 plus the cost of installation and replacement filters (additional annual cost estimate of $50–$120, depending on usage), for a total cost of $250–$320. The cost of these mitigation options could be prohibitive for some county residents. To make access to safe, clean drinking water more equitable, affordable nitrate mitigation resources should be made available and advertised to households in areas with nitrate well water levels ≥10 mg/L.

Considering the time, effort, and environmental impact of purchasing bottled water, the cost of installing and maintaining a point-of-use treatment system is the preferable option for households. The efficacy of a private well nitrate remediation program that would offer and aggressively advertise nitrate remediation options to homeowners with well water at or above the U.S. EPA nitrate MCL should be tested in an area that is experiencing nitrate contamination issues (like Eau Claire County). There is also a need for prioritizing education and outreach about the importance of monitoring nitrate levels in at-risk private wells (i.e., 5–9 mg/L nitrate).

Nitrate Contamination Risk Factors

The significantly higher average nitrate concentration in wells with agricultural indicators suggests agriculture is a source of nitrate contamination in private wells in Eau Claire County. Although studies have demonstrated that nitrate from septic system effluent is a contributor to poor well water quality (Shaw, 1994), our findings do not suggest septic systems are a significant source of nitrate in Eau Claire County. Other studies have also indicated that agriculture is the primary source of nitrate contamination compared with septic systems (Chern et al., 1999).

Casing depth was the only risk factor associated with elevated nitrate. Previous research indicates wells with casings less than 12.2 m (40 ft) have significantly more nitrate (Bundy et al., 1996), which is consistent with our study. Well age and depth had been previously identified as nitrate contamination risk factors but did not correlate with nitrate contamination in our study (Dakota County Environmental Management, 2003). The sandy soil, heavy agriculture, and thick sandstone aquifers allow for rapid and deep infiltration of water and water-soluble contaminants. This process and the increased likelihood of denitrification or lower nitrate concentrations in older groundwater at depth (Böttcher et al., 1990; Kraft et al., 2004) are the most likely explanation for higher concentrations of nitrate in wells with shallow casing.

Many of the wells for which records were available (n = 43) are constructed as open boreholes, with highly variable distances between the bottom of the borehole and bottom of the casing (0 up to 48.2 m [158 ft]) borehole depth below casing, median of 5.1 m (17 ft). This finding could explain the lack of correlation between nitrate concentration and well borehole depth in our study.

Conclusion

The frequency of samples with nitrate concentrations above the drinking water MCL in our study is similar to other regional studies where water quality was declared problematic. Agriculture appears to be the primary source of nitrate contamination of private wells in Eau Claire County. Solutions presented to resolve the nitrate problem in Wisconsin have traditionally focused on reducing nitrate fertilizer overuse on crops. Although this strategy is an important part of the solution, direct action is needed to protect homeowners from the adverse health effects associated with consuming water with nitrate ≥10 mg/L.

As most Wisconsin homeowners (70%) do not take action to reduce nitrate exposures from drinking contaminated well water (Knobeloch et al., 1997), local public health authorities must develop and implement interventions. Funds should be allocated to public health authorities in Eau Claire County or other areas experiencing similar nitrate contamination issues to promote and subsidize point-of-use drinking water treatment systems in homes with nitrate levels ≥10 mg/L. The efficacy of this approach could be studied as a pilot for other areas experiencing a similar rate of nitrate contamination in private well water. As casing depth was the only risk factor to have an association with nitrate contamination, private wells should be constructed with a casing depth greater than 12 m (40 ft) where possible to avoid infiltration of nitrate and other contaminants.

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