

Arsenic from community water fluoridation: quantifying the effect

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ABSTRACT

Community water fluoridation is a WHO recommended strategy to prevent dental carries. One debated concern is that hydrofluorosilicic acid, used to fluoridate water, contains arsenic and poses a health risk. This study was undertaken to determine if fluoridation contributes to arsenic in drinking water, to estimate the amount of additional arsenic associated with fluoridation, and compare this to the National Sanitation Foundation/American National Standards Institute (NSF/ANSI) standard and estimates from other researchers. Using surveillance data from Ontario drinking water systems, mixed effects linear regression was performed to examine the effect of fluoridation status on the difference in arsenic concentration between raw water and treated water samples. On average, drinking water treatment was found to reduce arsenic levels in water in both fluoridated and non-fluoridated systems by 0.2 µg/L. However, fluoridated systems were associated with an additional 0.078 µg/L (95% CI 0.021, 0.136) of arsenic in water when compared to non-fluoridated systems ($P = 0.008$) while controlling for raw water arsenic concentrations, types of treatment processes, and source water type. Our estimate is consistent with concentrations expected from other research and is less than 10% of the NSF/ANSI standard of 1 µg/L arsenic in water. This study provides further information to inform decision-making regarding community water fluoridation.

Key words | arsenic, drinking water, fluoride, water fluoridation

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INTRODUCTION

Community water fluoridation is the addition of fluoride to a public drinking water supply to prevent dental caries (McDonagh *et al.* 2000), a strategy that is recognized by organizations such as the World Health Organization (Petersen 2008), the Centers for Disease Control (Kohn *et al.* 2001) and Health Canada (Health Canada 2010). It is estimated that 40 countries have some type of artificial water fluoridation, with varying proportions of population coverage (Mullen 2005). According to Health Canada, the optimal concentration of fluoride in drinking water is 0.7 mg/L for communities that wish to fluoridate water for dental health (Health Canada 2010). The fluoride additives used to accomplish this may contain impurities, one of which is arsenic. These additives are subject to a system of standards, testing, and certification by the National

Sanitation Foundation/American National Standards Institute (NSF/ANSI), a non-profit, non-governmental organization. The NSF/ANSI standard 60 limits arsenic in fluoride additives to produce no more than 1 µg/L in water that has been treated to achieve 1 mg/L of fluoride (NSF International 2013). All drinking water systems in Ontario that fluoridate their water are required to use fluoride additives that meet NSF/ANSI standard 60.

In the province of Ontario, Canada, each municipality makes the decision about whether or not to fluoridate its water. Public concerns have resulted in some community drinking water systems discontinuing the addition of fluoride (Ontario Ministry of Health and Long-Term Care 2011). One of the debated concerns regarding community water fluoridation is that the hydrofluorosilicic acid used to

fluoridate water contains arsenic, and this poses a health risk to the population (Fluoride Action Network 2012).

Our search of the scientific and grey literature found five published studies that estimated the arsenic content of fluoride additives for drinking water (Weng *et al.* 2000; Casale 2001; Brown *et al.* 2004; Hirzy *et al.* 2013; NSF International 2013). Most measured the additives directly, and none measured the contribution of arsenic from fluoridation in actual drinking water systems. Most of the studies took measurements on a small number of samples, with only one study measuring more than 60 samples.

This study was undertaken to determine if fluoridation is a contributor to the concentration of arsenic in drinking water. In addition, we wanted to estimate the amount of arsenic that is added to water in actual drinking water systems as a result of fluoridation, using data gathered in Ontario, Canada, for systems supplied by ground or surface water. We also set out to see how this compares to previous estimates extrapolated from analysis of commercial fluoride products, and to the levels allowed under the NSF/ANSI 60 standard.

METHODS

Data sources

Publicly available data were obtained from the Ontario Ministry of the Environment and Climate Change (MOECC) Drinking Water Surveillance Program (DWSP) for the years 2005–2009 inclusive (<https://www.ontario.ca/environment-and-energy/drinking-water-surveillance-program-dwsp-data>). The DWSP is a voluntary water quality monitoring program that complements the regulatory monitoring of municipal systems in Ontario for scientific and research purposes (Ontario Ministry of the Environment 2013). Systems are chosen for participation based on multiple factors including the geographic location, surrounding land use, source water type, and type of treatment used. Drinking water samples collected by municipal partners are sent to accredited and licensed government laboratories for scientific analysis. Samples are tested for more than 270 inorganic, organic, and radiological water quality parameters. Results are stored electronically and transferred to a database where

they are subject to screening and verification to ensure quality controlled data. Once verified, the data are publicly released. Treatment process information was also obtained from the MOECC for each drinking water system, and this was linked to the DWSP sampling data using the DWSP ID. Any missing treatment and source water information was then obtained directly from the drinking water system report.

During this time period 8,756 sampling results of arsenic ($n = 5,218$) and fluoride ($n = 3,538$) concentrations in raw, treated and distribution water were obtained for 131 drinking water systems across Ontario. This included 1,998 raw water arsenic measurements, 1,610 treated water arsenic measurements and 1,610 distribution water arsenic measurements. Only raw and treated arsenic samples were used for this analysis. Raw water arsenic samples were matched with treated water samples that were taken on the same day or the day after the raw water measurement, within the same treatment plant. Data from matched samples were retained for analysis ($n = 1,517$).

Samples were excluded from the analysis if they contained a mixture of fluoridated and non-fluoridated water, or if their fluoridation status was unclear, to ensure the accurate categorization of fluoridation status. Similarly, individual treatment plants that had water drawn from both ground and surface water sources were excluded from the analysis, given that the proportion of ground and surface water mixing was unknown. One treatment plant was removed from the final analysis because the measured arsenic concentrations for that plant were extremely variable, and treated arsenic concentrations for the plant were unusually low in comparison to the raw water concentrations on the same day, given that the plant has no known treatment for arsenic. A sensitivity analysis was performed to determine the impact of this plant on the final results. After these exclusions, the final data set contained 1,329 paired raw and treated arsenic samples in 121 drinking water systems (Figure 1).

Relevant variables

The fluoridation status of each drinking water system was established by comparing fluoride concentrations in treated and raw samples. This was checked against the MOECC

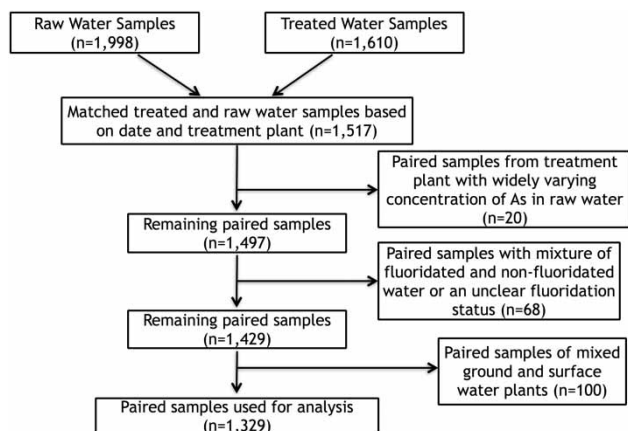


Figure 1 | Inclusion/exclusion logic diagram of paired samples used in this analysis.

classification of these same systems. Where there was disagreement between these two methods, the actual drinking water system report for the relevant plant was obtained to get the plant description and chemicals used. This report was then used to provide the final classification of fluoridation status.

The outcome variable was created by subtracting raw water arsenic concentrations from treated water arsenic concentrations for the matched samples. This difference was, on average, negative. Raw water arsenic concentrations were also included in the model to account for the baseline arsenic concentrations.

Treatment processes to be included as variables in the regression analysis were based on a review of processes that are capable of removing arsenic (McNeill & Edwards 1995; United States Environmental Protection Agency 2000; Chen *et al.* 2002; Health Canada 2006; Petrusevski *et al.* 2007). Those that were known to be capable of lowering arsenic concentrations and are used by drinking water systems in our data were included in the model. The processes included were coagulation, microfiltration and ultrafiltration.

The interaction between water source (ground vs. surface) and the presence of coagulation was included in the model. This was based on the information from our review of the arsenic removal process which stated that arsenic(III) is less efficiently removed by coagulation than arsenic(V) (United States Environmental Protection Agency 2000; Health Canada 2006; Petrusevski *et al.* 2007). Arsenic(III) is mainly found in ground water and arsenic(V) is the

main form found in surface water (Petrusevski *et al.* 2007), therefore coagulation is expected to have a greater effect on surface water systems than ground water systems.

Statistical analysis

R version 3.0.2 (R Core Team, 2013) was used for all analyses. Mixed effects linear regression using R package *lme4* (Bates *et al.* 2014) was performed. The random intercepts for treatment plants was used in this model and all other variables were included as fixed effects. To account for similarities in measurements within treatment plants, a mixed effect model was chosen because multiple samples were taken from each treatment plant. *P*-values were calculated using the likelihood ratio test.

T-test and chi-square tests were performed as part of the descriptive statistics on the main model variables to determine if they were significantly different in fluoridated and non-fluoridated systems.

RESULTS

The 1,329 samples used in the analysis were split nearly evenly between systems that add fluoride (49%) and those that do not (51%) (Table 1). Post-treatment samples taken from fluoridated systems had a mean fluoride concentration of 0.53 mg/L (SD 0.13 mg/L) compared to 0.12 mg/L (SD 0.21 mg/L) in non-fluoridated systems. Arsenic concentrations in the water entering the drinking water treatment system (raw) did not differ significantly by fluoridation status. Arsenic concentrations did decrease after treatment for both fluoridated and non-fluoridated systems; however, the amount of decrease was less in fluoridated systems (0.17 µg/L, SD 0.30 µg/L) than in non-fluoridated systems (0.22 µg/L, SD 0.32 µg/L). With respect to treatment, 83% of fluoridated systems used coagulation, whereas only 67% of non-fluoridated systems used coagulation and 3% of fluoridated systems used ultrafiltration while 10% of non-fluoridated systems used ultrafiltration. Seven percent of fluoridated systems had ground water as a source in comparison to 28% of non-fluoridated systems.

Using mixed effect linear regression, we modeled the impact of fluoridation status on the difference in arsenic

Table 1 | Descriptive statistics of study variables by fluoridation status. $n = 1,329$

Variable	Number (percentage) or mean (standard deviation)		
	Overall	Fluoridated	Non-fluoridated
Total sample	1,329 (100%)	653 (49%)	676 (51%)
Mean raw arsenic ($\mu\text{g/L}$)	0.67 (0.37)	0.69 (0.39)	0.66 (0.35)
Mean treated arsenic** ($\mu\text{g/L}$)	0.48 (0.31)	0.51 (0.33)	0.44 (0.28)
Mean treated-raw arsenic* ($\mu\text{g/L}$)	-0.20 (0.31)	-0.17 (0.30)	-0.22 (0.32)
Mean raw fluoride** (mg/L)	0.11 (0.19) ^a	0.07 (0.06) ^b	0.14 (0.25) ^c
Mean treated fluoride** (mg/L)	0.32 (0.27) ^a	0.53 (0.13) ^b	0.12 (0.21) ^c
Coagulation**			
Yes	994 (75%)	543 (83%)	451 (67%)
No	335 (25%)	110 (17%)	225 (33%)
Ultrafiltration**			
Yes	86 (6%)	19 (3%)	67 (10%)
No	1,243 (94%)	634 (97%)	609 (90%)
Microfiltration			
Yes	80 (6%)	35 (5%)	45 (7%)
No	1,249 (94%)	618 (95%)	631 (93%)
Source water**			
Ground	236 (18%)	47 (7%)	189 (28%)
Surface	1,093 (82%)	606 (93%)	487 (72%)

* $P < 0.0$.** $P < 0.001$ significantly different in fluoridated and non-fluoridated systems based on t -test or chi-square.^aMissing 139 samples.^bMissing 72 samples.^cMissing 67 samples.

concentrations between water entering the drinking water system and treated water (treated minus raw) while controlling for multiple factors (Table 2). Fluoridated systems were associated with an additional 0.078 $\mu\text{g/L}$ (95% CI 0.021, 0.136) of arsenic when compared to non-fluoridated systems ($P = 0.008$) while controlling for raw water concentrations, treatment processes (coagulation, microfiltration, ultrafiltration), and source water (ground vs. surface). Also, for every 1 $\mu\text{g/L}$ increase in raw water arsenic levels, the treatment process is associated with the removal of 0.55 $\mu\text{g/L}$ of arsenic on average while controlling for all other model variables.

Table 2 | Results of the mixed effect linear regression analysis

Parameter	Estimate (95% CI)	P-value
Fluoridation present	0.078 (0.021, 0.136)	0.008
Raw water arsenic (per 1 $\mu\text{g/L}$ increase)	-0.552 (-0.597, -0.506)	<0.001
Microfiltration present	-0.140 (-0.307, 0.027)	0.099
Ultrafiltration present	0.024 (-0.104, 0.153)	0.707
Surface water (ground as reference)	0.101 (-0.046, 0.248)	0.177
Coagulation present	-0.071 (-0.273, 0.130)	0.484
Interaction between Ground/Surface and Coagulation	-0.260 (-0.506, -0.013)	0.039

The outcome of the model is treated minus raw arsenic concentrations in $\mu\text{g/L}$. $n = 1,329$.

DISCUSSION

Our estimate of additional arsenic (0.078 $\mu\text{g/L}$) in fluoridated systems when compared to non-fluoridated systems is consistent with information found in different peer reviewed and non-peer reviewed sources. We used the results of five published studies and one unpublished study, which estimated the arsenic content of fluoride additives for drinking water (Weng et al. 2000; Casale 2001; Brown et al. 2004; Hirzy et al. 2013; NSF International 2013), to estimate the arsenic contribution to drinking water from fluoridation. Based on these studies, we would anticipate an increase in arsenic concentration of 0.004 $\mu\text{g/L}$ to 0.35 $\mu\text{g/L}$ in optimally fluoridated water as defined by Health Canada (see Table 3) (see supplementary material for original data and methods, available with the online version of this paper). The result we obtained is within this expected range, and likely provides a more extensive picture of what occurs in current day practice.

Long term exposure to arsenic in drinking water has been linked to cancer in humans (Morales et al. 2000). Standards to limit arsenic in fluoride additives (NSF International 2013) and drinking water (Health Canada 2006) have been developed to reduce the public health risks from arsenic exposures. Our estimate of additional arsenic is less than 10% of the NSF/ANSI standard of 1 $\mu\text{g/L}$ (NSF International 2013). This concentration of additional arsenic is also below the level of 'essentially

Table 3 | Expected arsenic in drinking water calculated based on levels of arsenic measured in the additive found in the peer reviewed and non-peer reviewed literature

Study	Number of samples	Arsenic concentration ($\mu\text{g/l}$) at 0.7 mg/L of fluoride in water ^a	
		Lower limit	Upper limit
NSF International (2013)	461	NA ^b	0.35
Brown <i>et al.</i> (2004)	4	0.025	0.162
Weng <i>et al.</i> (2000) ^c	Unknown	None given	0.168
Casale (2001) ^{c,e}	60 ^d	0.027	0.17
Ministry of Environment (Deshpande S., Personal Communication 2013)	8	0.004 ^f	0.018
Hirzy <i>et al.</i> (2013)	11	0.007 ^f	0.27

^aThis assumes that the raw water contains no fluoride, and that the concentration of 0.7 mg/L of fluoride is made up completely by the additive.

^bA lower limit is not appropriate since the method used in this study would result in arsenic concentrations below the level of detection.

^cArsenic content was from information provided by manufacturer.

^dThis number was not given explicitly in the paper, but inferred from the information given in tables.

^eAssumed in a 25% solution.

^fThis was not the lowest result found but rather the limit of detection.

negligible risk' of 0.3 $\mu\text{g/L}$, according to the Health Canada Guideline Technical Document for Arsenic (Health Canada 2006). The upper 95% confidence interval for the lifetime cancer risk associated with this 'negligible risk' concentration of 0.3 $\mu\text{g/L}$ is 1.9×10^{-6} to 1.39×10^{-5} (Health Canada 2006). Using the drinking water unit risk for oral exposure to arsenic from the USA EPA IRIS database, a lifetime cancer risk of 3.9×10^{-6} is estimated for a drinking water arsenic concentration of 0.078 $\mu\text{g/L}$ (Integrated Risk Information System 1995). All drinking water systems in this study were compliant with the Canadian drinking water guideline for arsenic of 10 $\mu\text{g/L}$ (Health Canada 2006), and the estimated amount attributable to fluoridation from this study is less than 1% of this guideline.

Factors other than fluoridation appear to account for a large portion of arsenic levels in the drinking water systems in our study. One important point to note is that arsenic concentrations decreased on average between raw and treated samples in both fluoridated (0.17 $\mu\text{g/L}$) and non-fluoridated

systems (0.22 $\mu\text{g/L}$) (Table 1), suggesting that treatment processes in Ontario are removing arsenic to some extent, even if they are not optimized to do so. Our model suggests that for every 1 $\mu\text{g/L}$ increase in raw water arsenic levels, the treatment process is expected to remove 0.55 $\mu\text{g/L}$ on average while controlling for all other model variables. Therefore, our findings suggests that the average removal efficiency of arsenic for the treatment systems in our study was 55%, after adjustment for fluoridation status, treatment type and source water.

We chose the difference between raw and treated arsenic concentrations as the outcome variable for several reasons. First, we are primarily interested in the impact that fluoridation has on treated water while accounting for the raw water concentrations going into the treatment plant. Also, treated water arsenic concentrations are not normally distributed, but the difference in arsenic concentrations between treated and raw water is normally distributed.

The results for one treatment plant were removed from the final analysis because the measured raw water arsenic concentrations for that plant varied by a factor of greater than 10. This was not found in any of the other plants, and suggests that the source water for this plant is different from the other plants in our study. A sensitivity analysis was performed to determine the impact of removing this plant on the results. It was found that the inclusion of this plant did not change the findings of this paper; however, the effect estimate for fluoridation does increase to 0.091 $\mu\text{g/L}$ and the *P*-value changes slightly to 0.039. This effect estimate still falls below the level of 'essentially negligible risk' from Health Canada (Health Canada 2006) and would correspond to a lifetime cancer risk of 4.6×10^{-6} using the IRIS drinking water unit risk (Integrated Risk Information System 1995).

This is the first published study, to our knowledge, that examines the effect of fluoridation on arsenic concentrations in actual drinking water systems. Studies have examined arsenic concentrations in the fluoridation chemicals themselves; however, we believe our method provides a more extensive picture of actual arsenic concentrations. Our estimate accounts for the effects of variation in source water quality, treatment process design and actual operations in over 100 systems. In addition, all drinking water systems do not fluoridate to the concentration of 0.7 mg/L recommended by Health Canada. The average concentration

of fluoride in our study was 0.53 mg/L in the treated water of fluoridated systems. There was also a natural background concentration of fluoride present in nearly all drinking water systems in our study, with an average raw fluoride concentration of 0.07 mg/L in fluoridated systems. Taken together, this suggests that the amount of fluoride that is actually added is less than would be estimated when the expectation is that all systems fluoridate to 0.7 mg/L and that the additive is the only source of fluoride.

There are also a number of limitations to our study. The DWSP sample is based on volunteer drinking water systems and therefore does not cover all of the municipal drinking water systems in Ontario and may or may not be a representative sample. However, systems are chosen for participation based on multiple factors including the geographic location, surrounding land use, source water type, and type of treatment used. In addition, water samples are collected by different people in each system and may be collected by different people within the same system; however, the sampling approach for the DWSP is standardized across the participating systems. The samples were also not analyzed to measure differences in particulate arsenic versus dissolved arsenic or different valence states of arsenic (As(III) vs. As(V)). To account for this, we controlled for many important factors including the treatment type, water source, and baseline raw water arsenic concentrations. Finally, our conclusions are limited to arsenic in fluoride additives that are NSF/ANSI certified, as required in Ontario. Given that the analysis was performed using Ontario data, caution must be exercised when results are generalized to other jurisdictions. Despite these limitations, our study is the first to our knowledge to look at the effect of fluoridation on arsenic concentrations in finished water across numerous drinking water systems, which can provide a more accurate estimate than measuring concentrations of arsenic in fluoridation chemicals.

CONCLUSIONS

The decision to add fluoride to a drinking water system or to stop fluoridating a system involves weighing a number of benefits, harms and costs. The potential health consequences from the unintended increase in arsenic is one consideration in such a decision. Our study provides

information for greater clarity on this issue to help inform decision making regarding community water fluoridation.

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CONFLICT OF INTEREST

The authors have no conflict of interest to declare.

DISCLAIMER

This work reflects the views of the authors and not necessarily those of Public Health Ontario or the Ontario Ministry of the Environment and Climate Change.

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