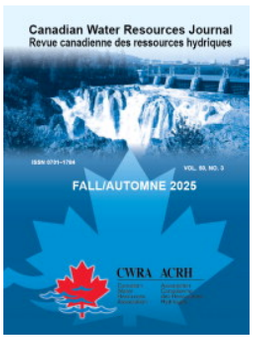

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Per- and polyfluoroalkyl substances (PFASs) contamination of groundwater in Canada: a (too) short review

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ABSTRACT

Concerns over per- and polyfluoroalkyl substances (PFASs) contamination of water continue to grow as more PFASs are found in more places and related guideline concentrations generally decline. Reports on the occurrence of PFASs in groundwater from around the globe have been published in scientific journals for over two decades now. Much of this work originates from the United States, China, and European countries. In this review, we investigated the state of studies publishing data on PFAS concentrations in groundwater or identifying PFAS sources to groundwater in Canada. We found and report on only 11 studies in scientific journals (by mid-2024), and the majority of these had linkages (direct or collaborative research, funding, or other key support) to federal or provincial governments. Potential reasons behind there being so few studies are discussed. Additionally, we pose and examine four key questions that highlight areas needing greater investigation in Canada. These are: (1) What is the state of PFASs in groundwater-sourced drinking water across the country? (2) What are background PFASs concentrations for groundwater? (3) What is the prevalence and distribution of PFASs sources to groundwater and risk posed by them? 4. How important is groundwater transport of PFASs to surface waters and aquatic ecosystems?

RÉSUMÉ

Les préoccupations relatives à la contamination de l'eau par les substances per- et polyfluoroalkylées (PFAS) continuent de croître à mesure que l'on trouve davantage de PFAS dans un plus grand nombre d'endroits et que les concentrations recommandées dans les directives diminuent de manière générale. Depuis plus de deux décennies, des rapports sur la présence de PFAS dans les eaux souterraines à travers le monde sont publiés dans des revues scientifiques. La plupart de ces travaux proviennent des États-Unis, de la Chine et de pays européens. Dans cette étude, nous avons examiné l'état des recherches publiant des données sur les concentrations de PFAS dans les eaux souterraines ou identifiant les sources de PFAS dans les eaux souterraines au Canada. Nous n'avons trouvé et rapporté que 11 études publiées dans des revues scientifiques, dont la majorité avaient des liens (recherche directe ou collaborative, financement ou autre soutien important) avec les gouvernements fédéral ou provinciaux. Les raisons potentielles de ce faible nombre d'études sont examinées. En outre, nous posons et examinons quatre questions clés qui mettent en évidence les domaines nécessitant des recherches plus approfondies au Canada. Ces questions sont les suivantes : 1. Quelle est la situation des PFAS dans l'eau potable provenant des eaux souterraines à travers le pays ? 2. Quelles sont les concentrations de fond des PFAS dans les eaux souterraines ? 3. Quelle est la prévalence et la répartition des sources de PFAS dans les eaux souterraines et quels sont les risques qu'elles présentent ? 4. Quelle est l'importance du transport des PFAS par les eaux souterraines vers les eaux de surface et les écosystèmes aquatiques ?

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Introduction

Per- and polyfluoroalkyl substances (PFASs), a group of many thousands of synthetic organic compounds containing at least one fully fluorinated methyl or methylene carbon atom, are commonly referred to as 'forever chemicals' because many of these compounds can persist in the environment. This is both because the C–F bond is extremely strong and stable (Smart 1994), and the fluorinated backbones tendency to adopt helical conformations allows the fluorine to provide shielding to the central carbon-carbon backbone (Mifkovic, Van Hoomissen, and Vyas 2022). PFASs were manufactured in bulk quantities as early as the 1930s (Hendricks 1953) and were present commercially by the 1950s (Buck et al. 2011) but have only received substantial attention as contaminants of emerging concern over the past few decades. Over this time, studies have revealed their widespread (global) occurrence in waters and soil (Kurwadkar et al. 2022; Salvatore et al. 2022), bioaccumulation in fish and other organisms (De Silva et al. 2021), and their potential to impair human and ecological health at very low concentrations (Sunderland et al. 2019). A few recent studies suggest that some PFASs deemed 'recalcitrant' may be susceptible to biotransformation (e.g. Huang and Jaffé 2019; O'Carroll et al. 2020). There are also a multitude of 'precursor' compounds (some also PFASs) that can degrade or transform in the environment into various (other) persistent PFASs (Key, Howell, and Criddle 1997).

The group of PFASs that has received the most attention to date for their presence in groundwater in many regions of the world are the perfluoro alkyl acids (PFAAs) (Grunfeld, et al. 2024), which include perfluoro carboxylic acids (PFCAs), such as perfluorooctanoic acid (PFOA), and perfluoro sulphonic acids (PFSAs), such as perfluorooctane sulfonate (PFOS). Depending on the number of carbons comprising the fluorinated chain, each of PFCA and PFSA have compounds considered long-chain ($>$C7 and $>$ C6, respectively), short-chain (down to C4), and ultra-short-chain (C1-C3) (Buck et al. 2011). Other PFAS groups that are becoming more commonly analyzed in groundwater include perfluoroalkane sulfonamides/sulfonamide ethanols/sulfonamidoacetic acids, per- and polyfluoroether carboxylic acids, ether sulfonic acids, and fluorotelemer acids (EPA method 1633, 2024; *via* liquid chromatography-tandem mass spectrometry). Various other types of PFASs (see Buck et al. 2011), some of which are found in aqueous film forming foam (D'Agostino and Mabury 2014; Patch et al. 2024a), could be investigated in groundwater with specialty expertise and equipment, namely high-resolution mass spectrometry.

PFASs are both hydrophobic and oleophobic, tend to reside at interfaces (surfactant properties), and display a strong resistance to heat and chemical attack. As a result of these properties, PFASs have been used in a wide range of consumer products (e.g. water- and stain-repellent materials {carpets, jackets, pyjamas, fast-food wrappings, etc.}, non-stick cookware, cleaning and personal care products, construction materials) and in firefighting foams designed for fuel fires (as in aqueous film forming foam; AFFF). Many pesticides and pharmaceuticals also contain PFAS. Principal (point) sources of groundwater contamination by PFASs include: (i) industrial facilities that have produced, processed, or used PFASs (Roberts 2025), (ii) places where AFFF (with PFASs) have been applied or stored (e.g. firefighter training facilities, military sites, airports, railroad crash sites, petroleum refineries, oil and gas extraction sites (Salvatore et al. 2022), (iii) solid waste storage sites (e.g. landfills), and (iv) wastewater treatment facilities (WWTF). Broader-scale PFASs sources have also been identified, including domestic wastewater septic systems (Subedi et al. 2015; Schaider, Ackerman, and Rudel 2016), application of wastewater-based biosolids to agricultural fields (Lindstrom et al. 2011), and atmospheric releases (Brandsma et al. 2019; Schroeder, Bond, and Foley 2021). As a result, low concentrations of PFASs are so prevalent in most environments as to be considered as background. Recently, the broad-scale and on-going accumulation of the PFAS trifluoroacetic acid (TFA) has been touted as a global threat (Arp et al. 2024).

Significant advancements have been made internationally in the understanding of PFAS fate and transport in the subsurface (see review by Hatton, Holton, and DiGuiseppi 2018). PFASs show strong retention in the vadose zone (above the water table) (Sharifan et al. 2021; Abraham et al. 2022), though many are quite soluble in water and will reach and travel through groundwater systems. Indeed, groundwater PFAS plumes can reach lengths of several km (e.g. Weber et al. 2017). Recent restrictions on the use of some PFASs (mostly long-chained PFAAs) in many countries around the world have led to greater use of short-chained PFASs (Dauchy 2019), which are typically more mobile in groundwater systems.

The primary motivation of most studies into PFAS contamination of groundwater is the concern for human health, with a focus on drinking or irrigation water from wells (e.g. Hu et al. 2016; Guelfo and

Adamson 2018; Kleywegt et al. 2020). In Canada, there is at present drinking water guidelines for PFOS and PFOA, and screening values for 9 other PFASs (Health Canada 2018a, 2018b, 2019). These are currently being reassessed (likely to be lowered), with a drinking water objective set at 30 ng/L for the summed concentrations (non-detect set as zero) of 25 PFASs recently announced (Health Canada 2024). There is also increasing interest in PFAS contamination of surface waters *via* the groundwater pathway, resulting in potential harm to fisheries and aquatic ecosystems while also threatening surface water sources of drinking or irrigation water. In Canada, only PFOS currently has a groundwater guideline for the protection of ecological receptors (7000 ng/L; CCME 2021), though a water quality guideline for the protection of aquatic life for PFOA is under development. PFAS water quality standards are rapidly developing across the world. These are lower in some jurisdictions than those in Canada (e.g. new US drinking water regulation of 4 ng/L for PFOS or PFOA; US EPA 2024) and are generally decreasing, especially for drinking water (Sauvé et al. 2023).

PFAS contamination of groundwater in Canada

In a recent global-scale review of scientific literature, Johnson et al. (2022) found 96 studies dating from 1999 to early 2022 that reported data on measured concentrations of PFASs in groundwater. These included relatively similar numbers of studies addressing primary contamination sites (e.g. manufacturing and industrial use, AFFF use), secondary contamination sites (e.g. landfills, sewage sludge application) and background sites (no known contaminant source), though favouring the primary sites. The first published study was by Moody and Field (1999) and described PFAS groundwater contamination at two fire-fighter training areas (FFTA) where AFFF was used. Over half of the studies have been published since 2015. The majority of studies came out of the U.S. (first) and China (second), both places where PFASs were manufactured and widely used in industry. While PFASs were not manufactured in Canada, most of the potential PFASs sources described above do occur in Canada. And yet, only three of the 96 studies were from Canada, leaving Canada tied for 8th (with Japan) in studies by country.

The earliest of the three Canada-based studies captured in the Johnson et al. (2022) review was performed by Meyer et al. (2011). Though largely focused on PFASs in surface waters, they also sampled groundwater seeps exiting from a shallow aquifer and a deep aquifer along the Scarborough Bluffs in Toronto, Ontario. There was no known PFAS source in the area (i.e. it was a background site), but it was an urban setting. From two sampling times, they reported concentrations of individual PFAAs < 5 ng/L in seepage from the upper aquifer and <1.5 ng/L in that from the lower aquifer.

The second referenced study from Canada was one by Martin et al. (2019) that was largely focused on analytical method development but employed samples from two airport sites (Newfoundland and Ontario). This has since been followed upon by the study of Liu et al. (2022), which considered these same two and an additional two (Ontario and Quebec) airports. Samples of soil (45) and groundwater (70) were assessed for the presence of many types of PFASs, including common targets (most of those found in US EPA method 1633) and others more rarely analyzed (including many precursors to PFAAs; and various cationic and zwitterionic PFASs). These studies noted that many of the less-frequently analyzed PFASs were found at non-negligible concentrations in groundwater, though PFOS was the dominant compound at all four sites. Across the sites, PFAS composition and spatial distribution were dependent on past AFFF use and the individual PFAS subsurface transport properties.

The third Canadian study captured in the global review, by Propp et al. (2021), looked for a variety of emerging contaminants, including a set of 17 PFASs, associated with historic landfills (i.e. those closed $>$ 25 years; many lacking liners or leachate collection systems). They collected 48 samples of leachate and/or (more often) leachate-impacted groundwater from the sites of 20 historic landfills in Ontario with closing dates from the 1920s to early 1990s. Elevated concentrations of PFASs were found for every landfill closed later than the 1950s, with highly variable concentrations and compositions between landfills. Further, PFASs concentrations at a few sites, even for some closed for $>$ 50 years, reached levels seen in modern landfills. These findings indicate that historic landfills can be important long-term sources of PFASs to groundwater.

Additional related studies for this review (completed in September 2024) were gathered through a literature search using i) search tools SCOPUS and Google Scholar, with a variety of PFAS-related search terms (e.g. 'PFAS', 'fluor'), and ii) citations of identified studies; similar to Johnson et al. (2022). One

earlier study that was not included in the Johnson et al. (2022) review was performed by Gottschall et al. (2017). They reported on 11 PFAAs, in addition to other groups of emerging contaminants, in soil, tile water, groundwater (2,4,6 m depths), and grain for a field receiving applications of municipal biosolids. A few of the PFAAs were detected at elevated concentrations in the tile drainage and shallow (2 m depth) groundwater, up to ~23 ng/L for PFOA, in comparison to the low to sub-ng/L concentrations in groundwater from reference plots. Another was a study by O'Carroll et al. (2020), which collected > 40 groundwater samples at a FFTA site in western Canada, with total PFAS concentrations (13 – 22 compounds) up to 3,500,000 ng/L reported. The study focused on differences in individual PFAS transport (sorption) and potential biotransformations of various precursor PFASs, with a strong focus on the microbial population.

Since the end of the Johnson et al. (2022) review period (approx. 2021), three additional studies reporting PFAS concentrations in groundwater from Canada have been published in the scientific literature (up to mid-2024); the Liu et al. (2022) follow-up study noted above and two that both included PFASs in drinking waters sourced by groundwater. The study by Kleywegt et al. (2020) included four drinking water systems supplied by groundwater out of 25 total drinking water systems tested in Ontario. PFASs concentrations (10 analytes) in treated drinking water sourced by groundwater were low (median = 3.8 ng/L) and significantly lower than those sourced by lakes and rivers ($p < 0.01$). Subsequently, Munoz et al. (2023) collected 463 tap water samples from 376 municipalities across Quebec for PFAS analysis. In line with the Ontario study, PFASs concentrations (sum of 42 analytes) was higher (by 12 X on average) in tap water produced from surface water (165 locations) than from groundwater (172 locations). However, 6 of the top 10 contaminated tap waters were sourced by groundwater. Note that a white paper published by the government of Quebec (MELCC 2022) around this same time included 76 samples from 33 drinking water systems supplied by groundwater. A similar finding for PFASs concentrations in supplies from surface waters versus groundwater was found.

The Johnson et al. (2022) review only captured published studies that reported field-based measurements of PFASs concentrations in groundwater. Our focus is slightly broader here to include studies that investigate potential PFASs sources to groundwater in Canada. Studies that only include theoretical or lab-based study of the fate, transport, or remediation of PFASs in groundwater are not considered. One such relevant PFASs source study was by Milley et al. (2018), who presented a decision-tree framework to identify airport/heliport sites in Canada potentially impacted by PFASs from AFFF releases at FFTAs and/or accident sites. Applying the framework, 152 (7%) of the 2071 airport/heliport sites investigated were deemed likely to have PFASs contamination. For a selected 25 sites of this group, PFASs contamination was confirmed by Canadian governmental departments at each of them. Additionally, for these 25 sites, the distance from release areas to the nearest surface water was calculated as within 2.5 kilometers for all of them, but within one kilometer for 19 sites. Thus, groundwater plumes from these sites pose a potential threat to nearby surface water ecosystems. Two additional PFASs source studies have reported PFASs in leachate from leachate collection systems from (presumed active) landfills in Canada. The first was by Li et al. (2012), which reported an average concentration (sum of 13 PFASs) of 2,950 ng/L in leachate from 28 landfills and dumpsites across Canada. A key finding was PFASs concentrations tended to be higher for landfills from southern Canada compared to those from northern (3 territories) Canada. The second study, by Gewurtz et al. (2013), reported concentrations of some individual PFASs in the 100s-1000s ng/L in leachate collected from 10 selected large municipal landfills across Canada. A key finding was that the summed concentration of the measured PFASs generally increased following on-site leachate treatment, presumably from the transformation of precursor compounds not captured in the analysis to PFASs that were analyzed.

Who is contributing to these Canadian studies?

The limited number of papers on PFASs concentrations in groundwater from Canada published over the last 25 years (i.e. since the first groundwater PFASs publication) suggests that this issue has not reached the levels of public or scientific concern as seems apparent in the U.S., China, and various places in Europe. But there may be other factors at play as well – perhaps financial, analytical, and related to available expertise.

While the number of journal-published studies detailed above is small (11), the researcher groups or institutions producing them are even more limited.

Three papers are products of a collaborative group from Université de Montréal and McGill University (Martin et al. 2019; Liu et al. 2022; Munoz et al. 2023). Another three were led or co-led by researchers from Environment and Climate Change Canada (ECCC), one with inputs from Ontario Ministry of Environment, Conservation and Parks (MECP), one in collaboration with University of Toronto (Meyer et al. 2011), and another in collaboration with McMaster University (Propp et al. 2021). Another study came out of the University of British Columbia (Li et al. 2012), though it was funded largely by ECCC, and had collaboration with the Department of Fisheries and Oceans (DFO). The study on biosolids application was led by Agriculture and Agri-Food Canada (Gottschall et al. 2017), with collaboration from the Ontario MECP. The Ontario MECP also led one paper on their own (Kleywegt et al. 2020). And finally, two papers involved researchers from Royal Military College of Canada (Milley et al. 2018; O'Carroll et al. 2020), the latter with collaborators from Australia.

The fact that the majority of these publications have direct ties to Canadian federal or provincial government researchers and/or directed funding indicates there is some interest in this topic by these government departments, all of which are involved with chemical contaminant policy and regulation. However, there still appears to be a lack of capacity or interest in this topic, especially by academics. The few numbers of research groups publishing this work likely reflects in part the limited number of institutions with internal capabilities to perform PFAS analyses, which requires expensive instrumentation and expert technical staffing. And where these capabilities exist, the PFASs research are often focused on (i) detections in other media (air, dust, precipitation, surface waters, soil), (ii) treatment or remediation of impacted soil or water, or (iii) toxicology or human health studies (based on personal experience of the authors). Having PFAS analyses performed at commercial labs is also very expensive on a per sample basis and can be limited to the common set of legacy PFASs. The widespread links to government funding or labs further supports the idea that this work has been limited by high financial and analytical requirements.

In the past, Canadian researchers have been at the forefront of study of other groundwater contamination, including petroleum hydrocarbons, chlorinated solvents, acid-mine-drainage, road salt, nutrients, and methane and brines from oil and gas development. However, the community of groundwater researchers in Canada is relatively small compared to the plethora of current issues (e.g. climate change, water supply withdrawals, oil and gas and mining development, non-point agricultural and urban pollution, groundwater-dependent ecosystems, etc.), so there may not be the capacity to take on PFASs in a major way. Furthermore, the nature of this topic generally requires expertise in analytical chemistry as well as hydrogeology, often necessitating interdisciplinary collaborations. These are not always easy to initiate or maintain, particularly beyond one or two close institutions.

Anecdotally, we know there is substantial work being performed across Canada by consultants for industry or government departments on their sites that have PFASs-impacted soil and groundwater, likely related to primary sources (e.g. AFFF use at airports, military bases and fire-fighter training areas). Some of this consultation may be done by or in collaboration with researchers. Assuredly, this work is generating new knowledge and understanding, some of which is shared through internal and external meetings, which is very valuable. But little of it is making its way to the scientific literature, probably for a variety of reasons. There may be little incentive or time for these investigators to draft a manuscript and see it through to publication. Also, there may be a confidentiality concern or a desire to 'keep a low profile' by the client that prevents or delays publication of study findings. Possibly some of the information obtained may not be novel enough (on its own) within the international literature to warrant publication in a scientific journal, noting that there are no national journals in Canada that focus on groundwater.

Interestingly, in a recent attempt to address limited publicly available data and uncover 'unseen science' on PFASs sites in Europe, Cordner et al. (2024) applied investigative journalism techniques (e.g. open-source intelligence (OSINT); freedom of information (FOI) requests) to better identify known and potential PFASs contaminated sites. The study identified 22,934 known contamination sites and 21,426 presumptive contamination sites across Europe, many more than expected. This added knowledge may drive new avenues of investigation into groundwater contamination by PFASs. Perhaps such an effort applied to Canada would work well. Although, Cordner et al. believe that PFASs sites were still missed in their investigation, due to a lack of geolocation, sampling, and publicly available data; limitations that may also apply in Canada.

Another option to estimate the threat of groundwater contamination from PFASs at a regional scale is to identify businesses or facilities that have potential

for PFAS use and contamination of groundwater. Ideally, this would include some assessment of the likelihood of contamination of each location. Such an effort is currently underway in Ontario, with Roberts (2025) recently (past our review window) identifying over 2000 industrial sites in Ontario which are likely to be either contaminated with or a source of PFASs. Many of these same sites were also identified as being located within 100 m of a groundwater supply well. Although the sites were identified, they have not been sampled.

What information is still missing?

Several of the aforementioned studies directly address the state of Canadian groundwater or the potential threat of PFASs to it (i.e. airports, landfills) at a regional or broader scale. Some of these also provide insights on PFASs occurrence or behaviour that are relevant beyond this country's borders. But there is still much we don't know about the state of PFASs in Canada's groundwater. Four key topics for which more information or understanding is clearly needed will be discussed in detail below. But first, a common issue with all of the groundwater PFAS data presented in the published studies discussed above deserves mentioning; that is, only a very limited number of PFASs in comparison to the many thousands that may be present in the environment are reported, and these are largely restricted to the common (legacy) PFAAs. For example, fluorotelomers are a set of PFAS that comprise a large proportion of PFAS used in products, but only a few of these are regularly analyzed (Grunfeld, et al. 2024). This is however complicated by the fact that telomers, especially telomer alcohols found in commercial products, readily transform in the environment (Dinglasan et al. 2004), suggesting both targeted and non-targeted analytical methods (high-resolution mass spectrometry, total oxidizable precursor assays, total organic fluorine) are required to best understand PFAS distribution and fate in the environment. This is not an issue unique to Canadian data, but rather it is a common problem globally that reflects the complexity and cost of PFAS analysis. In a review by Grunfeld, et al. (2024), they determined that for a global set of studies reporting on a total of 33,940 groundwater samples, the average analysis included only 16 distinct PFAS. However, in recent years, some researchers (globally) have analyzed groundwater for different types of underreported PFASs, like ultra-short-chain PFASs (Björnsdotter et al. 2019) and replacement compounds such as Gen-X (Brandsma et al. 2019). Though none to date have targeted the highly utilized fluorotelomer alcohols (Grunfeld, et al. 2024), although again this may be difficult as they quickly readily transform in the environment. Others attempt to detect as many compounds as possible with non-target analyses, such as Young et al. (2022) who identified 163 known PFASs and 134 previously unknown PFASs in an AFFF, and the recent study by Qi et al. (2024) that made use of an in-house PFAS database (collected from the NORMAN Suspect List Exchange and the US EPA CompTox Chemistry Dashboard) of 18,603 PFASs and non-PFAS fluorinated species and 6936 predicted MS/MS spectra, though not applied to groundwater samples. Another route to greater PFAS information gaining application is analytical measures to determine the total PFASs in a sample (see review of Nakayama et al. 2019), which are being continually developed (e.g. Tighe et al. 2021; Patch et al. 2024a,2024b).

What is the state of PFASs in groundwater-sourced drinking water across the country?

There are eight other provinces (aside from Ontario and Quebec), three territories and many First Nations – Metis – Inuit nations across Canada for which data on PFASs in groundwater-sourced municipal/community drinking water is not (openly) publicly available, let alone published in the scientific literature. For some of these, data may have been collected but not shared publicly (yet?), while others may be planning such data collection or be eager to collaborate on such efforts in future. The full extent of this data is not known. But even if such information was generated for each, there is no obligation to publish the data in the scientific literature or provide it publicly, and it would require a third party to assess the data at the national level and draw out insights that would be more broadly applicable. In the U.S., data on PFASs in groundwater-sourced drinking water have been captured under the federal (US EPA's) public water system monitoring initiative, the Unregulated Contaminant Monitoring Rule (UCMR). In the third round of this monitoring (UCMR3), six PFASs, including PFOS and PFOA, were included. This data set has been examined and reported in the scientific literature (Hu et al. 2016, Guelfo and Adamson 2018, Boone et al. 2019). Various states also provide publicly available data on PFAS sources or PFAS concentrations in groundwater (e.g. Michigan PFAS Action Response Team (MPART) – webpage and interactive

map; 2025). A listing with links to an additional 12 PFAS data portals from the United States, Europe, and Australia can be found in the Supporting Information. There is no regular monitoring program for drinking water or groundwater at the federal level in Canada and no easily accessible data from short-term monitoring projects. Thus, the overall extent or state of PFAS in much of Canada's municipal groundwater supplies is poorly known within the public realm.

Most Canadians reliant on private (groundwater) wells, which are most commonly used as a source for drinking water or irrigation water, likely have no real sense of the risk posed by PFASs, not knowing what the relevant sources are and how far they should be away from them. Several studies from the U.S. on known or suspected PFASs sources (i.e. manufacturing facility, land-applied WWTF biosolids) report PFAS concentrations for select private wells nearby (Hoffman et al. 2011; Lindstrom et al. 2011). These few results suggest that proximity to major PFAS sources is a key factor influencing PFAS contamination in private well water. But no widespread testing of private well water has been published to supplement these data. To the best of our knowledge there are no published data on PFASs concentrations in private wells in Canada.

What are background PFASs concentrations for groundwater in Canada?

Presumably there are no PFASs in deep, pristine aquifers filled with groundwater that is hundreds or more years old – that is unless we introduced it through drilling or sampling the well or by various other deep subsurface activities (d'oh!). But in more modern groundwaters, recharged since PFAS development, it seems rather a question of 'what are the PFAS concentrations?' rather than 'are PFASs there?'. Presumably nearly all modern groundwater has some PFASs given its ubiquitous use in human products and processes, its long-range transport across the globe (including TFA; Arp et al. 2024), and its incorporation into all aspects of the near-surface water cycle. What we might consider 'natural' sources to shallow groundwater could include dry or wet aerial deposition or recharging surface waters or managed aquifer recharge.

There are many questions related to this concept of background PFASs concentrations. Of most interest here is what range of PFAS concentrations should we expect to find in groundwater that is obtained from a typical municipal or domestic well, or that could be measured discharging to a surface water body, but that hasn't been significantly affected by a primary or secondary source of PFASs. And how do these concentrations vary with depth (reflecting time since surface exposure in some cases), between surficial and confined aquifers, across a watershed or region with its mix of historic and current land uses, and across the country? And how do we define the boundary between background and source-impacted groundwaters? As water quality guidelines continue to trend downward, particularly for drinking water, the derivation of background concentrations is likely to take on much greater significance. The authors are aware of at least one on-going international effort to derive a better understanding of background and ambient PFAS concentrations worldwide.

Some of the data collected on groundwater-sourced municipal water (Kleywegt et al. 2020; Munoz et al. 2023) likely represent background PFASs concentrations, though some of the municipal wells with higher concentrations were likely influenced to some degree by a PFAS source in the area. Additionally, so-called background wells have certainly been measured for comparison in many unpublished contaminated site studies. Thus, the potential for a substantial amount of background data is there, but in all of these surveys or studies, the well data will require close scrutiny to ensure no influences from primary, and especially, secondary PFASs sources.

What is the prevalence and distribution of the various known and possibly unknown PFASs sources to groundwater?

Many of the key PFAS sources likely to impact groundwater are known (or at least suspected) and have been reported on or at least mentioned in the literature (and in the introduction, here). These include primary sources like airports, military bases, and industrial facilities, and secondary sources like landfills, biosolids, agricultural runoff and wastewater lagoons. However, new PFAS sources are being highlighted in the literature all the time; one very recent example is textile mills (Dunn, et al. 2024). These are mostly point sources (i.e. contaminated sites), whereas non-point sources such as pesticide applications are seldom identified. Further, non-point secondary sources, which may have been downplayed in the past due to having lower source concentrations, such as biosolids application on agricultural fields (Lindstrom et al. 2011; Gottschall et al. 2017) and on-site wastewater treatment (septic) systems (Schaider, Ackerman, and Rudel 2016), may be driving broader-scale low-

level loading to groundwater, and subsequently to receiving surface waters. Of additional concern here is whether there are any sources that might be more prevalent or important in Canada that might not be captured or recognized by international studies.

Even if we know all the potential sources, there is then the business of identifying their locations within the various jurisdictions across Canada, at least for those deemed a likely threat. For clear targets like airports, this is a manageable task, though far from simple, as illustrated by Milley et al. (2018). It should be simple enough to identify all active landfill sites, but probably less so for historic sites (back to the 1950s). Likewise, active FFTAs should be known, but historical FFTAs or sites with AFFF application to large fires may be more difficult to locate. Identifying sites linked to large to small-scale industrial uses, such as auto manufacturing, pulp and paper, electroplating, textiles, and any place with large-scale fuel storage, would be more challenging. Some GIS-based work to map various PFASs point sources relevant to groundwater contamination has recently been performed and published for Ontario (Roberts 2025) and may be underway in other jurisdictions across Canada.

Even for identified contaminated sites, getting information on potential PFASs occurrence or use may not be straightforward. In a query of the Federal Contaminated Sites inventory, listing sites by 'PFAS' only recovered one site, which wasn't associated with groundwater impacts. Listing by 'Halogenated Hydrocarbon', which could be PFAS or could be chlorinated compounds, recovered 133 groundwater impacted sites; but combing this subset using the keyword 'PFAS' whittled this down to 29 sites (which is much more than one site). Additionally, the inventory listed 175 sites with a landfill, of which 76 reportedly had groundwater contamination of some kind. Though potentially not measured and largely not indicated, it's likely most of these 76 landfill sites (if open beyond the 1950s) have elevated PFASs concentrations in leachate-impacted groundwater.

Milley et al. (2018) stated, 'The scope of the issue regarding PFAS contaminated sites has not been evaluated in Canada to date. It is unknown how many sites may be contaminated, and therefore it is unknown what level of risk mitigation or clean-up is likely to be required into the future'. This is exacerbated by several factors. If it wasn't obvious by the nickname 'forever chemicals', we now know for certain that PFAS sources or sites created over half a century ago can still contaminate groundwater with PFASs substantially today (e.g. historic landfills in Canada, Propp et al. 2021; FFTAs, Ruyle et al. 2023). Also, as noted by Simon (2020), the application of 'low' drinking and groundwater standards for PFASs (parts per trillion) potentially leads to more 'sites' that must be considered. With the new Canadian drinking water objective for a group of 25 PFASs set at a combined concentration of 30 ng/L (Health Canada 2024), and with official guidelines currently in review and possibly to come out in a similar range, this may require a broader scope for identifying sources of PFASs groundwater contamination in this country.

How important is groundwater transport of PFASs to surface waters and aquatic ecosystems?

The study by Propp et al. (2021) included some shallow groundwater samples collected at the edges of streams, ponds, and lakes adjacent to several historic landfills, and these showed elevated PFAS concentrations. This indicates that leachate plumes in groundwater can be a source of PFASs to nearby surface waters. Otherwise, the papers listed above lack any investigation of groundwater-derived PFASs inputs to nearby surface waters. In fact, there are not many studies published in peer-reviewed journals on this topic regardless of the jurisdiction globally. Four such studies report on known contaminated sites. First, Tokranov et al. (2021) investigated PFASs transport and transformations in groundwater and surface waters in association with two flow-through lakes down-gradient of a historical FFTA and associated wastewater discharge basins. Focusing on pond sediments in plume discharge areas and groundwater recharge areas, they noted high PFAA precursor concentrations and summer-time losses, respectively, and highlighted the potential importance of dynamic biogeochemical conditions in controlling PFAA concentrations. Pétré et al. (2022) reported on PFASs concentrations and mass flux from groundwater to five tributaries of the Cape Fear River near a PFASs manufacturing facility in North Carolina (USA). Meanwhile, Divine, et al. (2023) described five example cases of where groundwater – surface water transfer of PFASs can be important, which included a variety of sources (FFTAs, WWTF, industrial wastewater discharges, airfield) and site conditions (most being contaminated groundwater impacting surface water bodies). And more recently, McFarlan and Lemke (2024) investigated a 3-km long PFASs plume emanating from an unlined historic landfill, with a numerical model predicting its discharge into several ephemeral ponds used to rear fish. Sampling found PFASs in the pond waters (up to

60 ng/L) and in several adjacent streams. In contrast, another study, by Briggs et al. (2020), used PFASs from an unknown source as one of several indicators of groundwater flow paths contributing discharge to a stream with Brook Trout spawning. These studies illustrate that groundwater inputs of PFASs to surface waters are occurring and thus may be important for mass loadings from various types of sites, though the findings are still limited.

Other studies infer mass loading of PFASs from groundwater based on concentrations measured in the receiving surface waters. For example, Ruyle et al. (2021) concluded from their study of Cape Cod watersheds in Massachusetts that 'legacy PFASs in slowly moving groundwater constitute a large source to the downstream coastal environment, representing a substantial lag between environmental PFASs releases and inputs to marine ecosystems'. For Canadian examples, groundwater transport and discharge to surface waters likely contributed to elevated PFASs concentrations found in (i) Etobicoke Creek downstream of the Lester B. Pearson International Airport, Toronto, Ontario (Moody et al. 2002; Awad et al. 2011), and (ii) the Welland River and Lake Niapenco downstream of the John C. Munro International Airport, Hamilton, Ontario (de Solla et al. 2012); both sites associated with AFFF contamination. However, no groundwater investigation has been published in either case.

PFASs contamination is widespread in Canadian surface waters (Scott et al. 2009; D'Agostino and Mabury 2017). The role of groundwater as a transport pathway between PFASs sources and these surface waters is generally not understood. This is illustrated in a recent study by Xia et al. (2024) that focused on air deposition of PFASs to the Great Lakes. As part of the work, they also produced a mass balance for PFBA (perfluorobutanoic acid), PFBS (perfluorobutane sulfonate), PFOS, and PFOA for the Great Lakes basin using additional data from previous studies (though notably limited). This included calculations of additional inputs from tributaries and WWTF, losses due to sedimentation in the lakes, and lake water transfer between lakes and out of the basin. They highlighted groundwater inputs of PFASs directly to the lakes as a gap in their balance that they were unable to determine. Some recent studies are beginning to consider this pathway, in calculating the distance between identified sources and nearby surface waters. For example, Milley et al. (2018) showed that there are 20 Canadian airports with verified PFAS contamination within 1 km of a surface water body (9 within 500 m). Likewise, Weber, Roberts, and Koch (2023) found that of the 1526 industrial sites in Ontario categorized as likely to contain or be a source of PFAS, 175 are within 1 km of a surface water body (114 within 500 m).

Finally, groundwater transport of PFASs to surface waters will lead to exposure to aquatic organisms living in the receiving and downstream environment, with potential bioaccumulation and toxicity impacts. Concentrations are likely to be greatest in the benthic zone, where impacted groundwater may be undiluted by the above receiving waters (Roy and Bickerton 2010), and may persist year-round. At the time of submission of this review, there were no published studies, from Canada or anywhere else, directed to aquatic exposure to PFASs through the groundwater pathway (although one, focused on two landfill sites in Canada, was just published while revising this manuscript (Roy et al. 2025)). As for studies addressing potential or actual impacts on aquatic organisms, there are few such studies regardless of the contaminant. In one of the few examples (and the only one from Canada), the recent work by Roy and Grapentine (2024) investigated the use of *in situ* cages placed at the sediment bed to assess potential impacts to benthic organisms exposed to discharging groundwater plumes. Two of the three sites investigated were historic landfills previously reported by Propp et al. (2021) to have elevated PFASs in the discharging leachate-impacted groundwater plumes. While there were indications of a toxic effect of the discharging plumes at both sites, it was not determined what role the elevated PFASs concentrations played versus those of the many other contaminants occurring in the leachate-impacted groundwater.

Conclusion

This review is relatively short. Clearly the scope of the review is quite narrow – one type of contaminant in one portion of the water cycle, focused on one single country. Also, remediation and prevention technology development and lab-based testing are excluded here, though such research is occurring in Canada (e.g. Duchesne et al. 2020; Abraham et al. 2022; Battye et al. 2022; O'Connor, et al. 2023; Rowe et al. 2023). But given (i) the globally-recognized concern for PFASs contamination, exemplified by further decreasing water quality guidelines, (ii) Canada's prevalent use of groundwater, which may even expand in a future quest for drinking water with ultra-low PFASs concentrations, and (iii) the historical strength of contaminant hydrogeology research in Canada, it feels like there should be more high-level

research papers to reference here. Perhaps some work is just slow to come out; indeed, we know of several potential papers in the pipeline. Perhaps more research groups and funding opportunities will join the effort soon, as new (likely lower) Canadian PFAS guidelines and/or screening values are released. In the least, hopefully this review makes it clear that there are numerous knowledge gaps in the literature which require the generation of data regarding PFASs presence in groundwater in Canada and the associated factors affecting their influence, transport and fate. And all of this data needs to be published.

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