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Ski wax use contributes to environmental contamination by per- and polyfluoroalkyl substances

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HIGHLIGHTS

- Per- and polyfluoroalkyl substances (PFAS) in ski wax abrade onto snow during use.
- Melted snow had the most PFAS, but soil and groundwater were also contaminated.
- Both long- and short-chain PFAS were detected.
- Ski waxes currently in use contain hazardous PFAS despite regulatory actions.

GRAPHICAL ABSTRACT

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are used in a wide variety of consumer products, including ski waxes, and are widespread persistent and hazardous environmental contaminants. We examined the environmental impact of ski wax use at an outdoor recreation area with significant cross-country ski activity by measuring PFAS levels in melted snow, soil and water following a collegiate ski race. We found extremely high levels of long- and short-chain PFAS (C4—C14) contamination in snow at the race start line ($\sum$[PFAS] 7600—10,700 ng/L), with the longer-chain analytes (C10—C14) predominating. The complement of 14 PFAS detected in snow matched what has been found in ski wax. This snow contamination was greatly reduced at a point 3.9 km into the race. Soil at the start line contained the four most predominant PFAS in snow at a mean individual concentration of 2.81 ng/g dry weight. Control soil contained only perfluorooctane sulfonic acid (PFOS), not found in other soil samples, at a concentration of 2.80 ng/g. Shallow groundwater from an on-site well contained only the shorter-chain PFAS (C4—C8), with a mean individual concentration of 4.95 ng/L. Our results suggest that ski wax use, from which fluorocarbons abrade at very high levels onto snow during a ski race, are the main source of PFAS contamination at our site. Regulation of ski wax use is warranted to reduce PFAS pollution.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) comprise a large family of industrial chemicals in widespread use in a range of products and applications (Sunderland et al., 2019). Their chemical properties, strong hydrophobicity, and stability confer high performance as water, grease, and stain repellants used in products like textiles, food packaging, fluoropolymer coatings, fire-fighting foams, and ski waxes. Among the most used and well-characterized PFAS are perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS).

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PFAS are characterized according to chemical structures and as short-chain or long-chain. PFOA is an example of a long-chain perfluorocarboxylic acid (PFCA), which has the structure F—$[CF_2]_n$—COOH where $n \geq 7$; for short-chain PFCAs, $n < 7$. PFOS is an example of a long-chain perfluorosulfonic acid (PFSA), which has the structure F—$[CF_2]_n$—SO_3H where $n \geq 6$; for short-chain PFSAs, $n < 6$.

Ski waxes are composed of hydrocarbons and fluorocarbons, and come in a variety of types (Plassmann and Berger, 2010). The exact composition of ski wax is rarely disclosed by manufacturers, but studies have detected high levels of long-chain fully fluorinated PFCAs and semi-fluorinated n-alkanes (SFAs) (Plassmann and Berger, 2010; Kotthoff et al., 2015). Fluorinated waxes are extremely hydrophobic and thus function to repel water away from the bottom of the skis, allowing for increased glide over the snow.

PFAS in products like ski waxes confer high-performance functions, but also have hazardous properties. Long-chain PFAS like PFOA and PFOS, in use for decades, are highly persistent, bioaccumulative and toxic chemicals (PBTs), and due to their widespread manufacture, use and disposal, are ubiquitous global pollutants. They are found at high levels in "hot spots" near local sources (e.g., 42,000 ppt and 205,000 ppt PFOS in groundwater near air bases in Michigan and Colorado, respectively, where firefighting foam was used) (Amon, 2019; Benesh, 2019), and at low levels elsewhere, including at low ppt levels in many municipal drinking water supplies (Evans et al., 2020; Safer States, 2020). PFAS are capable of long-range transboundary movement via air and water currents, and deposition in precipitation (UNEP, 2020). PFAS are detected in rainwater in the U.S. in areas near and far from known sources (NC DEQ, 2018; Ross, 2019).

The industrial applications, environmental fates, and toxicities of the thousands of PFAS other than PFOA and PFOS are not well characterized (Sunderland et al., 2019). Increasingly, as the hazards of PFOA and PFOS have become clear, short-chain PFAS and other fluorinated substitutes are being used, albeit without first studying their PBT properties. Emerging data suggest significant hazards of newer PFAS, including extreme persistence, a high degree of mobility in groundwater, long-range transport, wildlife contamination, and toxic effects (Brendel et al., 2018; Gomis et al., 2018).

Humans are exposed to PFAS because of their widespread and persistent environmental contamination and presence in our drinking water and food supplies (Sunderland et al., 2019). The largest U.S. biomonitoring study, conducted by the U.S. Centers for Disease Control and Prevention (CDC), has measured twelve PFAS in blood serum since 1999 and detected several in nearly every study participant. In 2016, the most recent year for which data are available, CDC detected five PFAS in people aged 12 and above: PFOS (geometric mean 4.72 μg/L), PFOA (1.56 μg/L), perfluorohexane sulfonic acid (PFHxS, 1.18 μg/L), perfluorononanoic acid (PFNA, 0.577 μg/L), and perfluorodecanoic acid (PFDA, 0.154 μg/L) (CDC, 2019). All but PFDA were also detected in children aged 3–11 at similar levels (CDC, 2019). Occupationally-exposed workers often have serum PFAS at extraordinarily high levels [e.g., 145–3490 μg/L PFOS and 72–5100 μg/L PFOA (Olsen et al., 2007)]. Body burdens are long-lasting in humans, with mean half-lives of selected PFAS in the range of 3–7 years in both workers and people with contaminated drinking water (Olsen et al., 2007; Li et al., 2018). Waxing skis has been shown to increase body burdens of PFAS in wax technicians (Freberg et al., 2010; Nilsson et al., 2010).

PFAS have a wide range of toxic effects in humans. PFOA exposure in humans is linked to kidney and testicular cancers (Barry et al., 2013), ulcerative colitis (Steenland et al., 2013, 2015), high cholesterol (Winquist and Steenland, 2014a), thyroid disease (Winquist and Steenland, 2014b) and pregnancy-induced hypertension (Darrow et al., 2013). Health effects in children include high cholesterol (Frisbee et al., 2010); lower levels of sex hormones, growth factors and thyroid hormone (Lopez-Espinosa et al., 2012, 2016); later age of puberty (Lopez-Espinosa et al., 2011) and attention-deficit/hyperactivity disorder (ADHD) (Stein and Savitz, 2011). The U.S. National Toxicology Program has classified PFOA and PFOS as immunotoxic (NTP, 2016). Research is lacking on the toxic effects of PFAS other than PFOA and PFOS.

PFAS are only weakly regulated under U.S. law, although PFOA and PFOS have been voluntarily phased out (EPA, 2020b). In 2016, the U.S. Environmental Protection Agency (EPA) proposed a non-enforceable health advisory level for PFOA and PFOS in drinking water of 70 ppt (EPA, 2020a). Several U.S. states have set binding standards for PFAS in drinking water at levels far below the EPA advisory (Safer States, 2020).

PFOA is listed on Annex A of the Stockholm Convention for global elimination (UNEP, 2020). The Stockholm Convention is an international treaty that regulates persistent organic pollutants (POPs). PFOS is listed on Annex B for global restriction of all uses except in fire-fighting foam, insect bait and metal plating (UNEP, 2020). The European Union (EU) has restricted numerous PFAS as "Substances of Very High Concern" under its chemicals law REACH ("Registration, Evaluation, Authorization and Restriction of Chemicals") (ECHA, 2020a).

Given the extraordinary hazards of PFAS, we asked if contamination could be detected at a local outdoor ski and recreation area and if ski wax use could be identified as a source. To our knowledge, this is the first report of PFAS contamination at a ski area in the U.S. (and anywhere outside of Sweden and Norway). Most environmental PFAS testing is focused on drinking water, but we wanted to know if snow, soil and groundwater were contaminated at this site.

PFAS contamination is common and widespread in Maine, as in many other regions, with numerous likely global and local sources. In the city of Waterville, Maine, where our study took place, sewage sludge is contaminated with PFOA and very high levels of PFOS, and the public drinking water supply and its source surface water contain low levels of three PFAS [PFOA, perfluoroheptanoic acid (PFHpA) and perfluorohexanoic acid (PFHxA)] (ME DEP, 2020). Comparing contamination at our site to other locations in Maine and elsewhere sheds light on this critical environmental health problem.

2. Materials and methods

2.1. Study area

Our study area was Quarry Road Trails, a public year-round outdoor recreation area in Waterville, Maine, a city of approximately 16,600 residents in central Maine. Quarry Road Trails has 10.4 km of groomed cross-country ski trails, in addition to trails for hiking, biking and snowshoeing, and a non-motorized boat launch on Messalonskee Stream, which forms the western border of the park. Due to its state-of-the-art snowmaking system, Quarry Road Trails sees a high volume of competitive and recreational cross-country skiing in the winter and is the host site for numerous regional, collegiate, high school and middle school ski races.

2.2. Sampling

Snow, water and soil samples were collected at several locations at Quarry Road Trails (Fig. 1). Frozen snow, run-off water and well water were collected on January 31, 2020, approximately 2 h after the completion of two freestyle cross-country ski races at the Eastern Intercollegiate Ski Association Colby Carnival event in which 163 skiers participated and fluorinated ski waxes were expected to be used. Soil samples were collected on May 14, 2020,

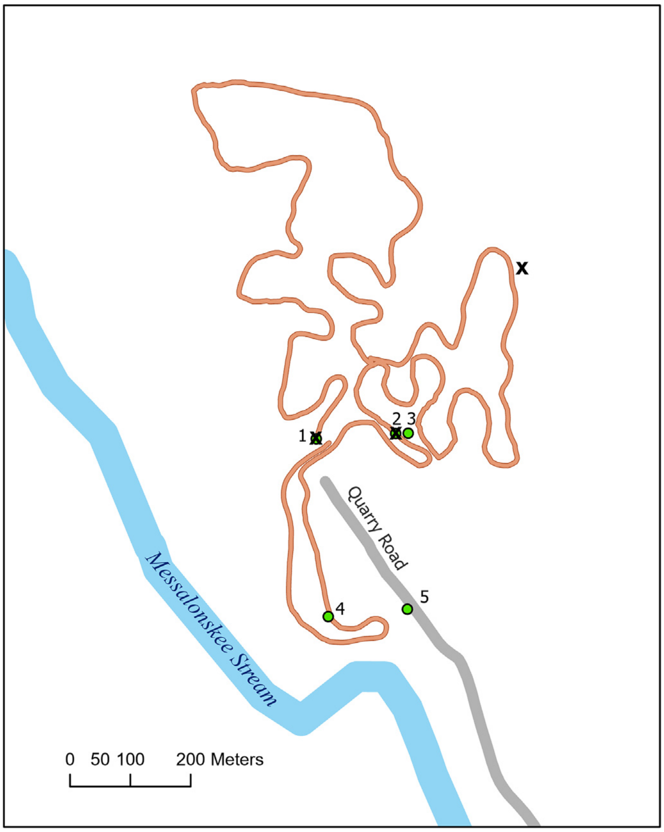

Fig. 1. Map of Quarry Road Trails in Waterville, Maine, showing the sampling sites, entrance road, ski race course, and nearby stream. (o) Melted snow or water sample site; (x) Soil sample site. 1, Start line of ski race course; 2, "Hero's Hill" on the ski trail; 3, "Hero's Hill" off the ski trail; 4, Run-off water site; 5, Shallow groundwater well.

after all snow had melted and the ground was thawed. All samples were collected in plastic 250-mL sample bottles provided by Alpha Analytical, Inc. (Westborough, MA). GPS coordinates for each sample site were taken in order to be able to gather soil from the same locations where snow was collected.

Two surface snow samples were collected at the start line of the ski race, one behind the start line and one over the start line, and another snow sample was collected on the race course on "Hero's Hill" 3.9 km away from the start line. A control snow sample was collected off the race course in untouched snow adjacent to "Hero's Hill." Snow was scraped to an approximate depth of 3–4 cm directly into bottles without using tools. Three bottles were filled for each of the four snow samples, but snow volume decreased significantly upon melting, so samples from each site were combined at the testing lab and analyzed as a single sample.

Run-off water was collected in duplicate from a gully adjacent to the ski race course, where water flows naturally from the property downhill toward Messalonskee Stream. This site was not near the start line area. Well water was collected in duplicate from a sink tap in a maintenance garage on site; water from this tap comes from a very shallow artesian well dug next to the garage. At each water and snow sampling site, a water field blank was collected.

Four surface soil samples were collected at the race start line area, and duplicate soil samples were collected on "Hero's Hill." A control soil sample was gathered in duplicate at the highest spot at Quarry Road Trails above and off the ski trail to minimize the possibility of chemical runoff from melting snow. Soil was scraped using a clean stainless steel spoon to a depth of approximately 3 cm, and mixed to homogenize the sample in a clean stainless steel bowl. For soil samples, equipment blanks were collected.

During sampling, care was taken to minimize contamination from sources of PFAS, including outdoor clothing and supplies (e.g., permanent markers, sticky notes, and recycled paper towels). A fresh pair of nitrile gloves was worn for each sample. All samples were stored on ice until they were picked up by staff of the testing laboratory, Alpha Analytical, Inc., at which point the chain-of-custody shifted to the lab.

2.3. *Chemical analysis*

Melted snow, water and soil samples were tested for the presence and concentration of 24 PFAS analytes in a panel created by Alpha Analytical, Inc. (Table 1) and using methodological guidance from the U.S. EPA (Shoemaker et al., 2008). At the testing lab, samples were spiked with surrogates, passed through a solid phase extraction cartridge, eluted with methanol, and cleaned up using GCB. Separation and identification of PFAS analytes was accomplished using a C18 liquid chromatography column and isotope dilution tandem mass spectrometry (LC/MS/MS). The concentration of each analyte was determined using the internal standard technique. Reporting limits (RLs) for liquid samples were 1.73–1.91 ng/L and for soil samples were 1.30–1.82 ng/g dry weight. Recoveries exceeded 50% for all analytes in liquid samples except PFOSA. Nearly all recoveries in soil samples collected on the ski trails exceeded 50%, and control soil recoveries were 20–65%. In all but the two positive soil samples from the start line, recoveries of PFTeA were very low (2–3% in start line soil without detections, 17% in "Hero's Hill" soil, and 0% in control soil), attributed to soil matrix effects.

Analyte concentrations reported here are taken directly from the laboratory reports. Concentrations of detected analytes in water and soil samples collected in duplicate are reported here as averages.

3. Results

3.1. *PFAS in snow*

We found PFAS contamination in all snow samples, some soil samples, and a well water sample from Quarry Road Trails (Table 2). Extremely high contamination was detected in snow at the start line, where skiers on freshly-waxed skis waited in line for their start time and then began the race. A snow sample collected just behind the start line ("Start line a") and another collected just over the start line ("Start line b") had the same detection pattern of 13 of the 24 PFAS included in the test panel. A 14th analyte, PFOS, was detected at a low level in "Start line a" snow only. All eleven perfluoroalkyl carboxylic acids (PFCAs) included in the test panel were detected, along with two fluorotelomers and one perfluoroalkyl sulfonic acid (PFSA).

The highest levels were detected for long-chain PFCAs with 10–14 carbons, with PFTeA (C14) found at by far the highest concentration (Table 2). Initial detections of some of the longest-chain PFCAs exceeded the laboratory's calibration range. The "Start line b" sample was diluted and re-run, but the "Start line a" sample was not due to limited sample volume; the level of PFTeA in this sample should be considered an estimate.

In "Start line a" snow, the order of analyte concentrations was PFTeA (C14) $\gg$ PFDoA (C12) > PFTrA (C13) > PFDA (C10) > PFUnA (C11) > PFOA (C8) > 6:2 FTS > PFHxA (C6) > PFHpA (C7) > PFNA (C9) > PFBA (C4) > PFPeA (C5) $\gg$ 8:2 FTS > PFOS. Concentrations ranged from 2.17 ng/L PFOS to an estimated 2760 ng/L PFTeA. The range of concentrations in this sample was too large to calculate a meaningful average. The summed concentration of all analytes

Table 1
PFAS analytes included in Quarry Road Trails sample testing.

PFAS Name	Abbreviation	CAS No.	Short-chain	Long-chain
PERFLUOROALKYL CARBOXYLIC ACIDS				
Perfluorobutanoic acid	PFBA	375-22-4	x	
Perfluoropentanoic acid	PFPeA	2706-90-3	x	
Perfluorohexanoic acid	PFHxA	307-24-4	x	
Perfluoroheptanoic acid	PFHpA	375-85-9	x	
Perfluorooctanoic acid	PFOA	335-67-1		x
Perfluorononanoic acid	PFNA	375-95-1		x
Perfluorodecanoic acid	PFDA	335-76-2		x
Perfluoroundecanoic acid	PFUnA	2058-94-8		x
Perfluorododecanoic acid	PFDoA	307-55-1		x
Perfluorotridecanoic acid	PFTrA	72,629-94-8		x
Perfluorotetradecanoic acid	PFTeA	376-06-7		x
PERFLUOROALKYL SULFONIC ACIDS				
Perfluorobutane sulfonic acid	PFBS	375-73-5	x	
Perfluoropentane sulfonic acid	PFPeS	2706-91-4	x	
Perfluorohexane sulfonic acid	PFHxS	355-46-4		x
Perfluoroheptane sulfonic acid	PFHpS	375-92-8		x
Perfluorooctane sulfonic acid	PFOS	1763-23-1		x
Perfluorononane sulfonic acid	PFNS	68,259-12-1		x
Perfluorodecane sulfonic acid	PFDS	335-77-3		x
FLUOROTELOMERS				
1H,1H,2H,2H-Perfluorohexane sulfonic acid	4:2 FTS	757,124-72-4	x	
1H,1H,2H,2H-Perfluorooctane sulfonic acid	6:2 FTS	27,619-97-2	x	
1H,1H,2H,2H-Perfluorodecane sulfonic acid	8:2 FTS	39,108-34-4		x
PERFLUOROALKANE SULFONAMIDES				
N-Methyl Perfluorooctane sulfonamidoacetic acid	NMeFOSA	31,506-32-8		x
N-Ethyl Perfluorooctane sulfonamidoacetic acid	NEtFOSA	4151-50-2		x
Perfluorooctane sulfonamide	FOSA	754-91-6		x

Table 2
Positive PFAS detections in melted snow, soil and water samples collected at Quarry Road Trails. ND, *Not detected at the reporting limit (RL) for the sample.* RLs, 1.73–1.91 ng/L for snow and water; 1.30–1.82 ng/g dry weight for soil. "*Start line* a" was just behind the start line; "*Start line* b" was just over the start line. "Soil 1" and "Soil 2" were two distinct soil samples (not duplicates) collected in close proximity at the start line; two other soil samples at the start line had no PFAS detections. Nine PFAS analytes for which there were no detections in any sample are not included in this table. Ten, 19 and 17 out of 24 PFAS tested for were not detected in any melted snow, soil or well water samples, respectively. All field blanks (snow and water) and equipment blanks (soil) were ND for all PFAS.

PFAS	Presence in ski wax (% ski waxes tested)[a]	Snow Start line a ng/L	Snow Start line b ng/L	Snow Hero's Hill ng/L	Control snow ng/L	Soil 1 Start line ng/g	Soil 2 Start line ng/g	Control soil ng/g	Well water ng/L
PFBA	65	97.1	311	ND	ND	ND	ND	ND	4.84
PFBS	38	ND	ND	ND	ND	ND	ND	ND	2.25
PFPeA	100	56.2	92.5	ND	ND	ND	ND	ND	5.66
PFHxA	88	255	427	ND	ND	ND	ND	ND	6.75
PFHpA	69	171	208	ND	ND	ND	ND	ND	4.53
PFOA	88	332	561	ND	ND	ND	ND	ND	6.62
6:2 FTS	–	297	98.6	ND	ND	ND	ND	ND	ND
PFOS	100	2.17	ND	ND	ND	ND	ND	2.80	4.00
PFNA	73	130	211	ND	ND	ND	ND	ND	ND
PFDA	88	580	1180	1.87	ND	ND	1.75	ND	ND
8:2 FTS	–	4.12	7.2	ND	ND	ND	ND	ND	ND
PFUnA	69	435	606	ND	ND	ND	ND	ND	ND
PFDoA	85	1530	1800[c]	3.74	4.08	2.82	ND	ND	ND
PFTrA	73	944	1000[c]	2.38	ND	3.61	ND	ND	ND
PFTeA	96	2760[b]	4210[c]	12.9	10.4	3.91	1.97	ND	ND

[a] Kotthoff et al. (2015).
[b] Initial detection exceeded calibration range; no dilution or re-assay done (value should be considered an estimate).
[c] Initial detection exceeded calibration range; ten-fold diluted sample re-assayed. RL = 18.2 ng/L.

detected was an estimated 7594 ng/L.

In "Start line b" snow, the order of analyte concentrations was similar but not identical: PFTeA (C14) ≫ PFDoA (C12) > PFDA (C10) > PFTrA (C13) > PFUnA (C11) > PFOA (C8) > PFHxA (C6) > PFBA (C4) > PFNA (C9) ≈ PFHpA (C7) > 6:2 FTS ≈ PFPeA (C5) > 8:2 FTS. Concentrations ranged from 7.2 ng/L 8:2 FTS to 4210 ng/L PFTeA. The range of concentrations in this sample was too large to calculate a meaningful average. The summed concentration of all analytes detected was 10,712 ng/L.

The complement of PFAS detected in snow at the start line was compared to PFAS known to be in ski waxes, which Kotthoff et al. (2015) found to be mostly PFCAs, with particularly high levels of PFOA (C8), PFDA (C10), PFHxA (C6), PFDoA (C12) and PFTeA (C14). Most ski wax samples they tested contained a nearly identical complement of PFAS as in our start line snow (Table 2). Our top four analytes, PFTeA, PFDoA, PFDA and PFTrA, were present in 96%, 85%, 88% and 73%, respectively, of ski wax samples tested by Kotthoff et al. (2015).

Snow from the ski trail on "Hero's Hill" had much less PFAS contamination (Table 2). The four PFAS detected at highest concentrations in start line snow were the only analytes detected in this sample, albeit at 270- to 470-fold lower concentrations. PFTeA was detected at the highest level (12.9 ng/L), followed by PFDoA (3.74 ng/L), PFTrA (2.38 ng/L), and PFDA (1.87 ng/L). The average concentration of individual detected PFAS was 5.22 ng/L, and the cumulative concentration of all four was 20.89 ng/L. No other PFAS analytes were found.

Our control snow sample from off the trail adjacent to “Hero’s Hill” had even lower PFAS detection (Table 2). Only two analytes, PFTeA and PFDoA, the predominant PFAS detected in snow, were found here, at similar concentrations (10.4 ng/L PFTeA and 4.08 ng/L PFDoA) as in “Hero’s Hill” snow from on the trail. The cumulative concentration was 14.48 ng/L.

3.2. PFAS in soil

Soil samples from the start line area were collected at four distinct spots in close proximity and two were found to contain the long-chain PFCAs most prominent in melted snow (Table 2). One positive sample had PFTeA, PFTrA and PFDoA at an average concentration of 3.45 ng/g dry weight. The other positive sample contained PFTeA and PFDA at an average concentration of 1.86 ng/g. For all four PFCAs in start line soil, the mean individual concentration was 2.81 ng/g. Cumulative concentrations in these two samples were 10.34 ng/g and 3.72 ng/g. The other two soil samples from the start line area had no PFAS detections, but they were wetter than the other two, and recoveries of PFTeA were very low, attributed to soil matrix effects.

Soil from “Hero’s Hill” had no PFAS detections. Control soil collected off the ski trail was contaminated with PFOS at 2.80 ng/g (Table 2). PFOS was not found in soil from the start line and was only found in one melted snow sample and at the lowest level of all analytes detected in snow. Recoveries of PFTeA were low in these soil samples.

3.3. PFAS in water

Shallow groundwater from a dug well was contaminated with seven of the 24 PFAS tested in this study, and the complement of analytes varied from that in our snow samples and in ski wax (Table 2). The order of detection level in well water was PFHxA ≈ PFOA > PFPeA > PFBA ≈ PFHpA > PFOS > PFBS. Five were PFCAs and two were PFSAs; all five PFCAs were the shortest chain chemicals tested (C4–C8), and the two PFSAs were C4 and C8. Longer-chain PFAS (>C8) were not detected in well water. The average concentration of detected analytes was 4.95 ng/L, similar to that in melted snow from “Hero’s Hill” and much lower than melted snow from the start line. The cumulative concentration of all detected PFAS in well water was 34.65 ng/L. In our run-off water sample, one duplicate had 2.90 ng/L PFTeA and no other detections; the other duplicate had no detections.

4. Discussion

4.1. PFAS in snow

To our knowledge, ours is the first study to document PFAS contamination at a ski area in the U.S. or anywhere outside of Sweden and Norway, and it adds to our understanding of the enormous global problem of PFAS pollution. We found some of the highest PFAS levels ever reported in melted snow, and ski wax use is the likely source, as the specific complement of PFAS in our snow samples matched that of PFAS detected in ski waxes. The chemicals found at the highest levels were long-chain PFCAs (C10-14), with PFTeA (C14) and PFDoA (C12) predominating. The major PFAS detected in glide wax powders were PFTeA and PFDoA, and PFTeA was the most abundant PFCA in the aerosol fraction of workroom air collected during ski waxing operations (Freberg et al., 2010). Thus, it is likely that the high contamination in our start line snow by these two PFAS, as well as other long-chain PFCAs, can be attributed to the skiers’ use of this type of wax. PFAS from ski wax readily abraded onto the snow surface at the start line, and we detected similar contamination in snow collected just behind the start line, where skiers lined up, and just ahead of the start line, where skiers began their sprint to start the race.

Our snow results are similar to those of the limited number of previous studies that have been conducted at ski areas. Snow was collected in 2010 at a ski area in Sweden that hosts the annual Vasaloppet cross-country ski race, and was found to have significant contamination by C8 and C14-20 PFCAs (Plassmann and Berger, 2013). Snow collected from cross-country ski wax testing tracks at a race site in Norway was highly contaminated with mostly long chain PFCAs (C12–C16), with the highest concentrations of PFTeA (1041 ng/L), PFDoA (491 ng/L), and PFTrA (137 ng/L) (Hanssen et al., 2019). These three PFCAs were also detected at the highest levels in our snow samples, suggesting that ski waxes with similar chemical compositions may have been used in both locations.

Summed PFAS concentrations in our two snow samples at the start line were extraordinarily high (an estimated 7594 ng/L and 10,712 ng/L) and exceeded levels reported in prior studies. Plassman and Berger (2013) reported $\sum$C6-22 PFCAs of 1400 ng/L; by comparison, our values of $\sum$C6-14 PFCAs were an estimated 7137 ng/L and 10,203 ng/L. Our study did not test for PFCAs with more than 14 carbons, but had they been included, we may have seen even higher cumulative concentrations of long-chain PFCAs. Hanssen et al. (2019) reported $\sum$C8-14 PFCAs of 1953 ng/L; by comparison, our values of $\sum$C8-14 PFCAs were an estimated 6711 ng/L and 9568 ng/L. PFTeA (C14) so greatly contaminated our samples that it was difficult to measure within the testing calibration range. The testing lab reported that ours were some of the highest levels of C12-14 PFCAs they had seen in aqueous environmental samples (P. Bassignani, pers. comm.).

Currently, ski teams are not required to disclose which ski waxes they use during races, so it is impossible to know what was used during the race that preceded our sampling. It is likely that fluorinated top coats were used, as is common in freestyle ski races. Until recently, these products have been ironed onto the ski surface to increase durability, but ironing creates PFAS fumes and thus is increasingly not allowed at competitive races, including at the race that preceded our sampling (EISA, 2020). It may be that by not ironing, these top coat waxes more easily rubbed off the ski surface onto the snow, resulting in the extremely high levels seen in our samples at the start line. It is interesting that the composition of the ski waxes used in this race did not appear to significantly differ from those used in Sweden and Norway up to a decade ago (Freberg et al., 2010; Plassmann and Berger, 2010). Ski wax products used in this race likely varied and may have included older products (T. Cote, pers. comm.). It was beyond the scope of this study to collect and test ski waxes for PFAS content.

In our study, PFAS contamination of snow decreased significantly several km into the race. High levels of eleven PFCAs were detected in snow at the start line, but low levels of only four PFCAs were detected on “Hero’s Hill,” 3.9 km from the start line. The four PFCAs in “Hero’s Hill” snow were the same four found at the highest level in start line snow, suggesting that snow contamination on “Hero’s Hill” also resulted from abrasion of ski wax chemicals during the race, but only residual wax was left on the skis by the time skiers got to this mark. Plassman and Berger (2013) also found that along a ski race course, PFAS levels in snow decreased rapidly with distance from the start line, with approximately 21-times less contamination 3 km into the race.

Our control snow sample collected in an untouched area off the ski trail had very low PFAS levels compared to the start line, and only the two PFCAs found at the highest levels in start line snow were detected here. It did not appear that skis had come into contact with this snow, but it is possible that ski wax chemicals abrading onto the snow surface nearby may have volatilized and deposited on adjacent snow or run off in melting snow from nearby trails, though not likely from “Hero’s Hill” because our control sample was collected above the trail. Deposition of PFAS-contaminated precipitation (rain or snow) may also have occurred, although the detection of analytes in control snow that were the most prevalent in other snow samples and in ski wax makes this explanation less likely.

4.2. PFAS in soil

In addition to snow, soil at Quarry Road Trails was also contaminated with PFAS that likely came from ski wax. Half of the soil samples taken from the start line area contained PFDA (C10), PFDoA (C12), PFTrA (C13) and/or PFTeA (C14), the most prevalent analytes in start line snow, at a mean individual concentration of 2.81 ng/g dry weight (parts per billion, ppb). In start line melted snow, the mean individual concentration of these four analytes was 1751 ng/L, equivalent to 1.75 ppb. We collected four samples at this site to increase our chances of gathering soil that had been directly underneath ski tracks. The two soil samples without detectable PFAS were wetter than the two positive samples, which may have diluted PFAS below the limit of detection or otherwise altered PFAS deposition, and soil matrix effects likely limited recoveries of the longest-chain PFCAs.

Soil collected on “Hero’s Hill” had no PFAS detections. Because snow from the ski trail at this site had much lower contamination by ski wax fluorocarbons, any deposition onto underlying soil may have been at a level below the limit of detection. A control soil sample only had PFOS, not detected in soil at the start line, at a level similar to the other PFAS detected in our soil samples (2.80 ng/g). In these samples, PFTeA contamination may have been possible but not detected due to soil matrix effects limiting recovery. It is unlikely that control soil was impacted by waxed skis, however, as it was gathered at the highest point in the park off and above the ski trail to minimize runoff and in a wooded area where it would be difficult to ski. As PFOS is a significant global contaminant prevalent in soil and other environmental matrices, atmospheric transport and deposition of PFOS may explain our control soil PFOS detection.

Our soil results are similar to soil monitoring in previous studies of ski areas in Sweden and Norway. Plassman and Berger (2013) reported soil contamination by PFAS, mostly C6-14 PFCAs, in the low ng/g range. Cumulative concentrations of $\sum$C6-22 PFCAs in their soil samples ranged from 0.059 ng/g (well below our limit of detection) to 19 ng/g dry weight. Our $\sum$C10-14 PFCAs in positive soil samples were 3.72 ng/g and 10.34 ng/g. Plassman and Berger (2013) found that soil samples collected away from the ski trail had fewer PFCAs and at lower concentrations than soil collected on the trail. At a ski center in Norway, soil was contaminated with mostly long-chain PFCAs in a pattern matching that of ski wax (Grønnestad et al., 2019). All individual PFCA concentrations were below 1 ng/g, lower than the concentrations in our positive soil samples. Soil samples from a reference site far from the ski area were contaminated with different PFAS, mostly PFOS and PFBA (Grønnestad et al., 2019).

Soil testing by Maine DEP at the Central Maine Regional Airport, a site 18 miles northwest of Quarry Road Trails that is not impacted by skiing activity, revealed a different pattern of PFAS contamination than in our start line soil. Testing of six airport soil samples for three PFAS analytes showed PFOA contamination at an average concentration of 0.62 ng/g and PFOS at 3.5 ng/g (ME DEP, 2020). PFOS was the only analyte detected in our control soil sample, but neither PFOA nor PFOS were in our start line soil samples, suggesting that sources of PFAS contamination differ between the airport and the ski race start line at Quarry Road Trails. Our limit of detection was too high to detect PFAS below 1 ng/g.

The fate of ski wax chemicals in melting snow and how they impact underlying soil cannot be predicted precisely. Certain PFAS are mobile in water and soil and may disburse over a large area during snow melt by running off in melt water, moving through soil, volatilizing, or being taken up by plants or wildlife (Plassmann and Berger, 2010; Grønnestad et al., 2019; Hanssen et al., 2019). In a laboratory simulation, Plassman et al. (2011) found that short-chain PFCAs partition in early meltwater fractions, and long-chain PFCAs partition in late meltwater and particle fractions that may be more likely to deposit on underlying soil. In general, decreasing water solubility is observed as PFAS chain length increases. It is possible that in our positive soil samples, contamination by only long-chain PFCAs can be explained by these distinct solubilities in melting snow, with short-chain chemicals dispersing in runoff and long-chain chemicals depositing in underlying soil. As the race start line was on an incline, snow melt likely led to some degree of downhill runoff of chemicals, away from our soil sampling area. Another explanation is that if most of the PFAS that contaminated the snow did deposit in soil, they did so at levels below the limit of detection, or they degraded over the 3.5 month interval between snow and soil sampling, although PFCAs and PFSAs, both long- and short-chain, are very persistent. Fates of individual PFAS vary, and some may be deposited repeatedly and persist from frequent ski wax use over long periods of time, even years.

4.3. PFAS in water

In our study, we detected PFAS in shallow well water at or exceeding levels seen in many drinking water supplies across the U.S. (Evans et al., 2020). Three of the seven PFAS in our well water sample are the same three PFAS that Maine DEP detected in the public drinking water supply in Waterville (ME DEP, 2020), but we found them at nearly double the concentration. Two additional PFAS in our well water sample were not detected in Waterville’s drinking water, and the final two detected in our sample were not tested for in Waterville’s water. The drinking water source for the city of Waterville is surface water from China Lake, 12 miles south of Quarry Road Trails and likely to have distinct contamination sources than shallow groundwater at our site. The PFOA + PFOS level in our well water was 10.62 ng/L (ppt), below the EPA health advisory level of 70 ppt but higher than the 3.80 ng/L detected in Waterville’s drinking water (ME DEP, 2020). The cumulative PFAS concentration in our well water was fairly high at 34.65 ng/L, and the PFOA level of 6.62 ng/L (ppt) exceeds the health-based PFOA drinking water threshold of 1 ppt recommended based on child immunotoxicity (Grandjean and Budtz-Jørgensen, 2013). This well water is considered non-potable and is not believed to be ingested by users of Quarry Road Trails, but it is used for hand-washing and other cleaning activities, and unforeseen ingestion may result in significant PFAS exposure, especially in children.

Because our well water sample contained only short-chain PFCAs and PFSAs plus PFOA and PFOS, but not the longer-chain fluorocarbons that predominate in start line snow and ski wax, it may be that well water contamination is not due to ski wax use, but rather to an unidentified local and/or global source. If ski wax use is the source, it seems that only the shorter-chain but not longer-chain components of ski wax move through the environment at our site to such an extent as to contaminate shallow groundwater at a site far from the start line area. Short-chain PFAS have been shown to be more mobile in water and soil than long-chain PFAS, less likely to bind to sediment or other particulate fractions, and more likely to contaminate drinking water and be subject to long-range aqueous transport (Brendel et al., 2018).

Other possible localized sources of well water PFAS include snow melting off the ski trail grooming equipment, which is stored in a garage adjacent to the well, although the full complement of ski wax fluorocarbons that might be picked up by the groomer from snow on the ski trails was not detected in the well water. Another possibly relevant source is emissions from high-performance outerwear, boots and other gear worn by visitors to Quarry Road Trails that are coated with a waterproof fluorocarbon-based treatment. Many PFCAs and PFSAs, along with fluorotelomer alcohols, are detected at varying concentrations in coated jackets (Dreyer et al., 2014), but it is not known how much may be released into the outdoor environment when these articles are worn.

In Maine, land-spreading of sewage sludge, often contaminated with PFAS from municipal waste and/or industrial sources, is common (Maine PFAS Task Force, 2020), and sludge is often mixed into commercially-available compost. We do not know if sludge or compost has been spread at Quarry Road Trails, and historic data on PFAS contamination is lacking because PFAS have only recently been recognized as priority hazardous pollutants and begun to be detected widely. A 2006 study in Germany determined that surface water and groundwater impacted by land-spreading of PFAS waste along the Ruhr River were contaminated with the identical complement of PFCAs and PFSAs as in our well water sample (Skutlarek et al., 2006); they tested for but did not detect long-chain PFAS other than PFOA and PFOS. A farm in Maine known to be impacted by PFAS from sludge spreading (Maine PFAS Task Force, 2020) had groundwater contamination by similar PFAS and at similar levels as in our well water (ME DEP, 2020).

In surface waters, the ratio of PFHpA to PFOA is used to indicate if the likely source of contamination is local (PFHpA:PFOA<1) or atmospheric (PFHpA:PFOA>1) (Simcik and Dorweiler, 2005). It is not known if this ratio can also be used for groundwater, but if so, our PFHpA:PFOA ratio of 0.68 suggests a local PFAS source. Whether ski wax use, sludge spreading or other source is responsible for groundwater contamination at Quarry Road Trails is unknown, but this contamination is concerning as it matches what is seen at known impacted sites in other parts of the world.

4.4. Regulation of PFAS in ski wax

The use of fluorinated ski waxes in competitions has recently been restricted. The International Ski Federation (FIS) announced a ban on fluorinated ski wax use in international cross-country ski competition starting in the 2020-21 season (FIS, 2020). The Norwegian Ski Association, Nordiq Canada, and several U.S. states, including Maine, have banned at least some types and uses of fluorinated ski waxes (Rathke, 2020). The New England Nordic Ski Association has restricted some uses of fluorinated products and acknowledges that “the wax industry will shift as the international market responds to mounting regulation of ingredients in Europe” (NENSA, 2019).

The global elimination of PFOA under the Stockholm Convention took effect in 2020 (UNEP, 2020), which means that C8-based fluorinated ski waxes will no longer be produced or sold, and many fluorinated ski waxes will likely be C6-based (Howard, 2020). As most ski wax manufacturers are located in countries that are part of the EU and are parties to the Stockholm Convention, they must follow EU laws and treaty obligations. The U.S. is one of very few countries not a party to the Stockholm Convention (UNEP, 2020), so the global elimination of PFOA technically does not apply in the U.S., even though it will impact ski wax imports. Germany and Sweden are developing restrictions for C9–C14 PFCAs (major components of fluorinated ski waxes) under EU’s REACH, and these chemicals may also be considered for global regulation under the Stockholm Convention (ECHA, 2020b). PFHxS has already been recommended for inclusion in the Stockholm Convention (UNEP, 2020).

The U.S. lags Europe in PFAS regulation, but the EPA is currently developing a PFAS Action Plan (EPA, 2020b). Numerous U.S. states have PFAS restrictions pending, notably bans on PFAS-containing consumer products (though not ski waxes), and several are setting enforceable drinking water standards (Safer States, 2020). Maine’s PFAS Task Force made numerous recommendations for state-level action on PFAS, including requiring manufacturers to disclose intentional use of PFAS in consumer products and to use safer alternatives when possible, which in theory could apply to ski wax. As a consumer product, ski wax may be one of the more dangerous PFAS sources because people can be in direct contact with the fluorocarbons in wax, powder and liquid forms, and these chemicals are readily released from the product during use.

For now, many fluorinated waxes remain on the market. One of the leading ski wax manufacturers is Norway-based Swix, which, according to its website, sells products ranging from a “100% fluorocarbon” glide wax powder to the SwixPro™ “fluoro-free” waxes (Swix, 2020) being rolled out this year in time for the FIS ban. Steve Poulin, CEO of Swix Sport USA, claims that the company “is going to push for a fluoro-free environment” (Howard, 2020). Italy-based Star Ski Wax is selling a new glide wax that, according to its website, “uses a new generation of fluorocarbon that is already compliant with the new European regulations of 2020” (Star Ski Wax, 2020). Specific chemical information is not disclosed by either of these manufacturers.

Ski wax may be one of the first categories of PFAS-laden products to become PFAS-free, but this will depend on current and future restrictions on ski wax use and regulations of fluorinated chemicals. Given the widespread persistence and toxicity of PFAS, certain EU countries (Germany, the Netherlands, Norway, Sweden and Denmark) are spearheading data collection to support restricting PFAS as an entire class, rather than on a chemical-by-chemical basis that would take far too long to regulate in order to protect people from harm. The European Chemicals Agency acknowledges that for PFAS, “a holistic group approach to the regulatory assessment and risk management needs to be explored” (ECHA, 2020b). This approach makes sense in light of the difficult burden of PFAS remediation, which is prohibitively expensive and in some cases impossible once PFAS are released into the environment (Kwiatkowski et al., 2020). Widespread global contamination, human exposure and dangerous toxicities warrant a precautionary approach to PFAS regulation.

The main limitation of our study is that we were not able to test more samples due to high laboratory costs, but this does not change our conclusions that PFAS contaminate Quarry Road Trails and that ski wax use is likely the main source of snow and soil contamination on the ski trails, and possibly also of shallow groundwater, though other local sources for groundwater contamination should be considered. A future study could investigate additional locations at our study site, and at various times throughout the year. It might also be useful to study ski wax use in competitive versus recreational skiing as well as the efficacy of emerging ski wax use restrictions to reduce environmental contamination.

5. Conclusions

Continued use of fluorinated ski waxes is a significant source of environmental contamination at Quarry Road Trails and likely at other cross-country ski areas in the U.S. and around the world. Human PFAS exposure is probable at these sites due to ski waxing activities and contact with contaminated snow, soil and water. Also, even though ski wax use may not be one of the top contributors to global PFAS pollution, it should not be overlooked as a global source because of the long persistence of its fluorocarbon components.

To protect human health and the environment from PFAS contamination, it is prudent to continue developing safer practices and policies for ski wax use. We should eliminate fluorinated ski wax production and sale, monitor ski wax use at races to ensure compliance with restrictions, and ensure that people are aware of the risks and are able to minimize their exposure to PFAS-containing products and contaminated snow, soil and water. We should also set careful guidelines for the disposal of existing stocks of fluorinated ski waxes to prevent further environmental emissions. Generally, we should continue testing for environmental contamination and PFAS body burdens in humans, expand research into the health impacts of low and high exposures to older and newer PFAS, strengthen regulations of PFAS as a chemical class, and invest in research to develop truly safer alternatives.

Credit author statement

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Declaration of competing interest

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