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Levels, Patterns, and Biomagnification Potential of Perfluoroalkyl Substances in a Terrestrial Food Chain in a Nordic Skiing Area

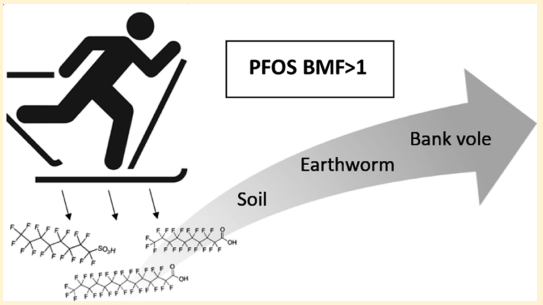
Randi Grønnestad,*[,†] Berta Pérez Vázquez,[†] Augustine Arukwe,[†] Veerle L. B. Jaspers,[†] Bjørn Munro Jenssen,[†] Mahin Karimi,[‡] Jan L. Lyche,[‡] and Åse Krøkje[†]

[†]Department of Biology, Norwegian University of Science and Technology (NTNU), NO-7491 Trondheim, Norway

[‡]Department of Food Safety and Infection Biology, Norwegian University of Life Sciences (NMBU), NO-0033 Oslo, Norway

Supporting Information

ABSTRACT: Perfluoroalkyl substances (PFASs) are used in a wide range of consumer products, including ski products, such as ski waxes. However, there is limited knowledge on the release of PFASs from such products into the environment and the resultant uptake in biota and transport in food webs. We investigated levels, patterns, and biomagnification of PFASs in soil, earthworms (*Eisenia fetida*), and Bank voles (*Myodes glareolus*) from a skiing area in Trondheim, Norway. In general, there was higher PFAS levels in the skiing area compared to the reference area with no skiing activities. The skiing area was dominated by long-chained perfluorocarboxylic acids (PFCAs, ≥70%), while the reference area was dominated by short-chained PFCAs (>60%). The soil PFAS pattern in the skiing area was comparable to analyzed ski waxes, indicating that ski products are important sources of PFASs in the skiing area. A biomagnification factor (BMF) > 1 was detected for Bank vole$_{whole}$/earthworm$_{whole}$ for perfluorooctansulfonate in the skiing area. All other PFASs showed a BMF < 1. However, it should be noted that these organisms represent the base of the terrestrial food web, and PFASs originating from ski wax may result to higher exposure in organisms at the top of the food chain.



INTRODUCTION

Perfluoroalkyl substances (PFASs) are ubiquitous and persistent anthropogenic chemicals in the environment.[1] They are a group of surface-active compounds that are applied in a wide range of consumer products, such as textiles, carpets, impregnating agents and in some types of ski products, such as ski waxes, gliders, and powders.[2] The global production of ski waxes is estimated to be several tons per year.[3] During the last decade, the production and use of ski waxing products have increased considerably, and the chemical composition of these products is continuously evolving.[4] In cross-country and downhill skiing, these products are applied to increase performance, as the fluorinated molecules enhance the glide on the water film between the ski and snow surface.[5] However, abrasion of these products from the ski sole results in deposition of the PFASs to the nearby environments.[6] Because PFASs are very persistent, they can remain in the environment for decades, creating PFAS-hotspots in the skiing areas.[7] However, little is known about the environmental levels of PFASs in these areas and their uptake in biota and transport in food webs.

In recent times, there has been an increasing focus on PFASs in consumer products, their toxicity, persistence in nature, and potential spread to the environment.[2,8,9] Particularly, the two most toxic congeners, namely, perfluorooctanoic acid (PFOA) and perfluorooctansulfonate (PFOS), have received much attention. In the year 2000, the US Environmental Protection Agency (USEPA) banned PFOS, and in May 2009, it was added to Annex B of the Stockholm Convention on persistent organic pollutants (POPs: www.pops.int). In 2010, the maximum content of PFOS allowed in products was reduced to equal or below 10 mg/kg in the Commission Regulation (EU) No. 757/2010.[10] In Norway, the use of PFOS was banned in firefighting foams, textiles, and impregnation agents (max. content 0.005%) in 2004 (FOR-2004-06-01-922, 2004). In addition, a maximum content of 0.1 mg/kg PFOS is allowed in other types of products (FOR-2009-06-22-827, 2009). A similar restriction for PFOA is under development. However, several PFASs continue to be manufactured as the industry has not yet found suitable replacements for these compounds.

According to Kotthoff et al.,[2] ski waxes had the highest concentrations of both perfluorocarboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs), compared to a wide range of other consumer products. Despite the legislative focus, PFOA and PFOS were the main contributors to total PFAS levels in most of the consumer products.[2] Studies on blood serum from professional ski waxing technicians have shown elevated concentrations of PFCAs, compared to the general population.[5,11] This is of great concern, since studies have shown that

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PFASs can, among others, lead to several adverse health effects, such as hormone imbalance,[12,13] immune suppression,[14,15] and alterations of lipid homeostasis.[16,17] This has resulted in an increased focus on the levels and possible effects of PFASs on human health.[11,18,19] However, there is limited or no data regarding the effects of PFASs on wildlife species inhabiting areas where these products are being used and released into the environment.

Herzke et al.[20] reported high PFAS levels in earthworms in skiing areas in Oslo, Norway, compared to a reference site. Although only five samples were used for this study, these results gave reason for concern and follow-up studies were recommended. Furthermore, snow chamber studies revealed that PFCAs elute in concentrated peaks from the melting snow,[21] potentially affecting biota during their most vulnerable stage of development in the spring.

Therefore, the aims of the current study were (1) to investigate the levels and patterns of PFASs in a Nordic skiing area in different environmental matrices, including soil, earthworms (*Eisenia fetida*), and Bank voles (*Myodes glareolus*), and to compare these levels and patterns to a reference area with no skiing activities and (2) to investigate to which extent these contaminants biomagnify in the food chain. These data will be useful in regulatory aspects of PFASs in ski wax, providing better insights into the sources and exposure routes in the environment.

MATERIALS AND METHODS

Study Area. The study area was "Granåsen skisenter" (63°22′N, 10°18′E), located approximately 10 km from the Trondheim city center (Norway, Figure S1 in the Supporting Information). Granåsen is the main arena for winter sports in Trondheim and hosts an annual ski jumping World Cup event in addition to a range of other regional, national, and international competitions in cross-country skiing. Thus, Granåsen offers several cross-country ski tracks that are used for training and competitions by amateurs and hobby skiers.

As a reference site, a natural forest area not used for ski-sports was chosen in the vicinity of an ecological farm next to Lake Jonsvatnet (63°20′N, 10°33′E). This site is approximately 15 km away from Trondheim city center (Figure S1 in the Supporting Information). The lake supplies drinking water to the Trondheim and surrounding communities. The two study areas have quite similar vegetation, consisting of mainly mosses and different species of *Ericaceae*.

Study Matrices. Chemical analyses of soil are useful for detecting the concentration of contaminants in the environment,[22] and earthworms (*E. fetida*) are considered one of the most suitable model organisms for monitoring and assessing soil pollution as they are integral soil macroinvertebrates.[23,24] Earthworms constitute an important part of the diet of local rodent species and serve as the gateway for chemical movement from the contaminated soils into the terrestrial food web. Thus, earthworm was chosen as a test organism because of its critical role at the base of the investigated terrestrial food web and its constant contact and ingestion of soil. Earthworms are susceptible to chemicals, providing information on the bioavailability of soil contaminants.[24] We chose Bank voles as a model organism because it is an important intermediate species in the terrestrial food chain, being preyed upon by raptors and carnivorous mammals,[25] and feeds on roots, seeds, buds, and berries, in addition to earthworms and other invertebrates. In addition, they have a relatively small home range, so we could expect that their contaminant levels are representative of the area where they were caught.

Sampling. The soil samples were collected in June 2017 and 2018 from the Granåsen and Jonsvatnet areas. The upper layer (constituting 3−10 cm depth and an area of approximately 1 m^2) of soil was collected and dried (40 °C for 48 h). Five samples per year in Granåsen and Jonsvatnet were chosen for chemical analysis. Only soil from locations where both earthworms and Bank voles had been sampled were selected for analysis.

The sampling of earthworms was performed in June of 2018 by digging 5−10 cm into the soil, using a metal spade and collecting the animals in sealed plastic bags. They were immediately frozen at −80 °C until analysis. The short time between collection and freezing did not allow them to empty their guts, as this would be more representative of how they serve as Bank voles' prey.

The catching, handling, anesthesia, sampling, and euthanizing of the Bank voles were approved by the Norwegian Food Safety Authority (Mattilsynet; references no. 2017/76552) and by the Norwegian Environmental Agency (Miljødirektoratet; reference no. 2017/4061). Permissions for the collection of Bank voles were also given by the land owners. The sampling and handling were performed in accordance with the regulations of the Norwegian Animal Welfare Act and EU legislation (3R). The collection of Bank voles was performed in June 2017. All traps were live traps of type "Ugglan" baited with rye bread dipped in sunflower oil and peanut butter (all food products were sold as "ecological food material"). The Bank voles were sacrificed by cervical dislocation. The animals were weighed, measured, and sexed. The livers were dissected and snap-frozen in liquid nitrogen and stored at −80 °C. The eyes were dissected for later age determination and stored in 10% formalin. In total, 21 and 31 individuals were caught at the Granåsen and Jonsvatnet areas, respectively. For more details on sampling and handling, see the Supporting Information.

Age Determination. The age of the Bank voles was determined using the weight of the dried eye lenses.[26] The lenses were dried at 80 °C for 24 h, and each lens was weighed to the nearest 0.1 mg. The mean weight of the two lenses was used to calculate approximate age. The calculations of age were done according to Kozakiewicz,[26] using the following formula: $Y = 0.013x + 4.610$, where y = lens weight and x = age (days).

Because the growth rates of the eye lenses are larger during the first three months of their life,[26] the Bank voles which were estimated to be less than 3 months old were recalculated using the following formula: $Y = 0.063x + 1.050$.

Chemical Analysis. The PFAS concentrations were analyzed at the Environmental Toxicology Laboratory, Norwegian University of Life Sciences (NMBU), Oslo, Norway. The analytical procedure of PFASs is described by Grønnestad et al.[27] The samples were analyzed for the following PFASs: ten PFCAs: perfluorobutanoic acid (PFBA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUdA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), and perfluorotetradecanoic acid (PFTeDA), three PFSAs: perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), and PFOS, and five perfluoroalkane sulfonamide derivatives (FASAs): perfluoro-1-octane sulfona-

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mide (FOSA), *N*-methyl perfluoro-1-octane sulfonamide (*N*-MeFOSA), *N*-ethyl perfluoro-1-octane sulfonamide (*N*-EtFOSA), 2-(*N*-methyl perfluoro-1-octane sulfonamido) ethanol (*N*-MeFOSE), and 2-(*N*-ethyl perfluoro-1-octane sulfonamido) ethanol (*N*-EtFOSE).

Extraction of Biota. 0.5 g of Bank vole liver or whole earthworm was weighed for chemical analysis. Brief description of the method is as follows: internal standards (^{13}C-labeled equivalents, 20 ng/mL; Wellington Laboratories, Table S1 in Supporting Information) were added prior to double extraction with methanol. Cleanup was accomplished using active carbon (EnviCarb). See more detailed description in Supporting Information.

Extraction of Soil Samples. The dried soil sample (5 g) was weighed for the chemical analysis. The method for soil extraction was similar to that of biota; however, an additional step with addition of 2 mL of 200 mM sodium hydroxide (NaOH) prior to the extraction and 200 μL of 2 M hydrochloric acid (HCl) after extraction was included in the procedure.

Analysis. The final extracts were analyzed by separation on high-performance liquid chromatography with a Discovery C18 column (15 cm × 2.1 mm × 5 μm, Supelco, Sigma-Aldrich, Oslo, Norway), connected to a precolumn; Supelguard Discovery C18 column (2 cm × 2.1 mm × 5 μm, Supelco, Sigma-Aldrich, Oslo, Norway). Detection and quantification were accomplished with a tandem mass spectrometry (MS−MS) system (API 3000, LC/MS/MS System). The injected volume was 5 μL.

External standards were used to produce a standard curve from which the PFAS levels were calculated, using the instrument control and data processing program Mass Hunter Quantitative analysis Version B.05.02 (Agilent Technologies). The limits of detection (LODs) were calculated as 3 × SD of the procedural blanks (see blank values Table S5 in the Supporting Information), and the limits of quantification (LOQs) were calculated as 10 × LOD. Where no blanks were detected, LOQs were determined as 10 × signal-to-noise ratio (S/N). For the soil and earthworm samples, individual LOQs were determined for each sample because of matrix effects. Further information on the chemical analyses and LOQs can be found in the Supporting Information (Tables S1−S4).

Quality Assurance. The Environmental Toxicology Laboratory is accredited by the Norwegian Accreditation as a testing laboratory according to the requirements of NS-EN ISO/IEC 17025 (TEST 137).

For each series of maximum 30 samples, three blank samples, one blind, and four recovery samples were run. Mean of procedural blanks, consisting of internal standards and solvents, was subtracted from each series separately because of variation between series. The relative recovery rate in Bank voles ranged from 84 to 128% for the PFCAs, 78−129% for the PFSAs, and 86−115% for the FASAs. For the earthworm samples, recoveries ranged from 110 to 140% for the PFCAs, 99−115% for the PFSAs, and 106−141% for the FASAs. For the soil sample, recoveries ranged from 91 to 140% for the PFCAs and 97−124% for the PFSAs. It was not possible to analyze the FASAs in the soil samples because of poor response of the internal standards.

Contaminants with concentrations above LOQ in more than 50% of samples were included in the statistical analyses, and missing values (i.e., <LOQ) were assigned a random value between the LOQ and zero.

Calculations of the Biomagnification Factor. Because bioaccumulation of PFASs are highly tissue and substance specific,[28] the most appropriate approach for calculating biomagnification factors (BMFs) is to use whole-organism concentrations for both predator and prey.[29] In the earthworms, whole-body concentrations were analyzed. However, in the Bank voles, only liver concentrations were analyzed, and the liver mass accounted for 5−7% of the total body mass. Thus, to provide indications on the potential of PFASs to biomagnify at the base of a terrestrial food chain, individual whole-body concentrations were calculated for the Bank voles and used for estimation of BMFs. Because the PFAS concentrations generally are higher in liver tissue than in other tissues,[30] we assumed that the PFAS concentrations in the rest of the tissues on average were 10% of that in the liver. This was based on calculations of whole-body concentrations of PFASs in mice.[31] Whole-body concentrations were thus estimated as C_{whole} = (liver fraction × C_{liver}) + (fraction of other tissues × C_{liver} × 0.1).

The BMF was calculated as the ratio between Bank $vole_{whole}$/$earthworm_{whole}$ for individual PFASs at the Granåsen and Jonsvatnet sites of values above LOQ.

Statistical Analysis. The program R (version 3.5.3, the R project for statistical computing) was used for the statistical analysis. Normal distribution was tested with Shapiro Wilk's test, and homogeneity of variance was tested with Levene's test. Data were log-transformed prior to data analyses to reduce deviation from normality and homogeneity of variance. Two sample Student's *t*-tests were used to test for significant differences between the skiing and reference areas. The significance level was set at 0.05, and all tests were two-tailed.

There was no significant difference in PFAS concentrations between years for soil samples (*t*-test, $p = 0.1$ for Granåsen and $p = 0.09$ for Jonsvatnet), so the 2017 and 2018 samples were pooled for statistical analysis. There was no effect of gender (*t*-test, $p = 0.7$ for Granåsen and 0.6 for Jonsvatnet) or age (*t*-test, $p = 0.8$ for Granåsen and $p = 0.3$ for Jonsvatnet) on Bank vole liver PFAS levels; therefore, the contaminant data were not separated into subgroups for statistical analysis.

RESULTS AND DISCUSSION

PFASs in Soil and Earthworms. There was no significant difference (*t*-test, $p = 0.8$) in the mean-summarized PFAS concentrations (ΣPFAS) in the soil samples from the Granåsen skiing area and the Jonsvatnet reference area (Figure 1a), showing concentrations of 1.57 and 1.54 ng/g d.w. (dry weight), respectively. In the earthworms (Figure 1b), the mean ΣPFAS levels were 35% higher at Granåsen than Jonsvatnet (10.5 and 6.92 ng/g w.w. (wet weight), respectively). However, this difference was not significant (*t*-test, $p = 0.08$) due to large individual variation (see Table S3 in the Supporting Information).

For the PFCAs, PFDA was the most predominant compound in the soil samples from Granåsen, while the long-chained PFTeDA was the most predominant compound in the earthworms from Granåsen. At Jonsvatnet, PFBA was the dominating compound in both soil and earthworm samples. A study from the Antarctic Peninsula found that PFBA was found in 80% of lichen samples,[32] indicating that PFBAs are present in quite pristine areas. For the PFSAs in both soil and earthworms, PFOS was the dominating compound, representing a significant portion of the PFSA group. The FASA derivatives were FASA derivatives were

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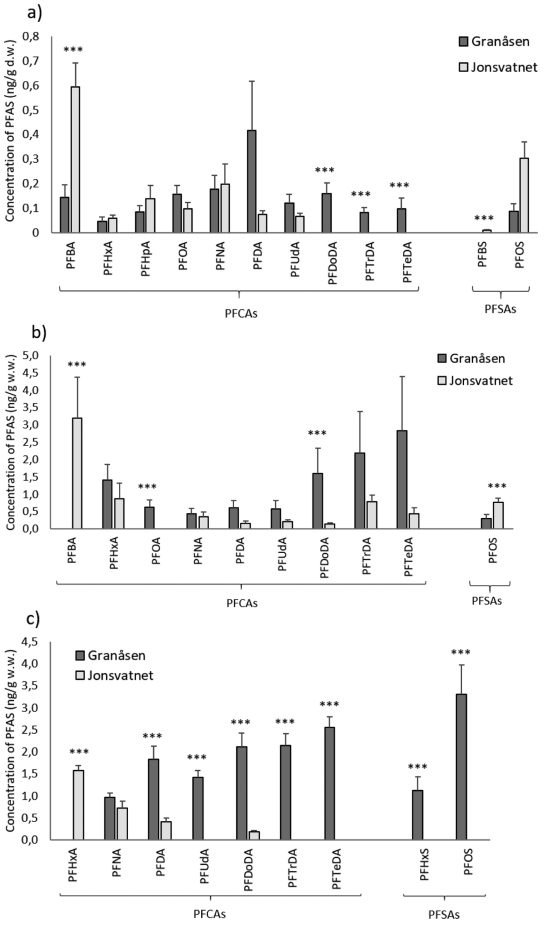

Figure 1. PFAS concentrations in (a) soil (ng/g d.w.) during 2017 and 2018 at Granåsen (n = 10) and Jonsvatnet (n = 10), (b) earthworms (ng/g w.w.) during 2018 at Granåsen (n = 13) and Jonsvatnet (n = 13), and (c) Bank voles (*M. glareolus*) during 2017 at Granåsen (n = 21) and Jonsvatnet (n = 31). Error bars indicate standard error of the mean (SE). Asterisks (*) indicate significant site differences (t-test); * = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$. PFASs with missing bars have levels <LOQ.

below LOQ in earthworms in both study areas (see Table S3 in the Supporting Information) and could not be analyzed in soil samples.

Concentrations measured in soil were below 1 ng/g d.w. for all individual PFASs at Granåsen (Figure 1a). This concentration is low, compared to similar soil studies in other areas near skiing tracks.[20,33,34] The ΣPFAS levels in soil from a skiing area in Oslo, Norway, was 10.3 ng/g d.w. in 2016[33] and 7.1 ng/g d.w. in 2017,[34] compared to 1.57 ng/g d.w. at Granåsen. According to the Norwegian guidelines on classification of environmental quality of soil, concentrations of 100 ng/g d.w. of PFOS represent the threshold for clean soil (FOR-2004-06-01-931, § 2, attachment 1). This indicates that in both the skiing area in Trondheim and Oslo, the levels are several orders of magnitude below the threshold for contaminated soil.

For the earthworms (Figure 1b), the PFAS concentrations at Granåsen were below the concentrations reported in the Oslo skiing area.[20] The ΣPFAS concentrations in earthworms from Granåsen, 10.5 ng/g w.w., were lower than the concentrations reported in Oslo, where concentrations ranged from 34.8 in 2015 to 70 ng/g w.w. in 2017.[20,33,34] Recently, an LC_{50} (lethal concentration at which 50% of the population is killed) of approximately 478 mg/kg was reported for PFOS in earthworms,[35] and this LC_{50} value is several orders of magnitude above the levels measured in the present study (<0.011 mg/kg). Nevertheless, there are potential and other severe effects, besides mortality, that can be observed at lower PFOS concentrations. For example, Zheng et al.[35] reported DNA damage in earthworm coelomocytes at their lowest test concentration of 50 mg PFOS/kg. Elsewhere, Xu et al.[36] observed that exposure to soil PFOS concentration of 10 mg/kg (their lowest test concentration) produced DNA damage and oxidative stress in earthworms. Therefore, although the individual PFOS concentrations reported in earthworms from skiing areas in Norway are below concentrations that produce acute toxicity (i.e., mortality), it is not possible to conclude on other long-term chronic effects. In addition, we must consider mixture toxicity scenarios, which might lower the toxicity thresholds.

PFASs in Bank Voles. To our knowledge, there are no previous studies of PFAS levels in Bank voles at skiing areas.

The mean ΣPFAS concentration was 5.7 times higher in Bank voles from Granåsen, compared to Jonsvatnet (15.6 ng/g w.w. and 2.74 ng/g w.w. at Granåsen and Jonsvatnet, respectively, Figure 1c). This difference was statistically significant (t-test, p = 0.02). There was no difference in the sex ratio (F/M = 33/67 at Granåsen and 34/66 at Jonsvatnet) or age distribution (t-test, p = 0.2) between the two areas. Thus, the differences in PFAS concentrations between the two areas are not caused by differences in these biological factors. The FASA derivatives were below LOQ in both areas (see Table S4 in the Supporting Information).

PFAS levels in Bank voles from forest and subalpine biotopes in Sweden have previously been reported.[37] However, that particular study was not linked to skiing areas, and mean concentrations of PFAS in the biotopes varied from 5.8 ng/g w.w. to 18.7 ng/g w.w., with the highest concentrations in Våladalen.[32] It should be noted that there are skiing areas in Våladalen (https://www.valadalen.se/en/cross-country-skiing), but no information is provided on the exact sampling locations of the voles in relation to these skiing areas.[32] Several studies have reported PFAS levels in terrestrial animals, however, these are mainly from areas near factories, where PFASs are produced or used.[38,39]

The concentrations of the long-chained PFCAs (C10−C14) were significantly higher in Bank voles from Granåsen compared to Jonsvatnet (t-test, PFDA: $p < 0.001$, PFUdA: $p < 0.001$, PFDoDA: $p < 0.001$, PFTrDA: $p < 0.001$, PFTeDA: $p < 0.001$, Figure 1c), while no difference between the two areas was observed for PFNA (C9, t-test, p = 0.25). For the short-chained PFHxA (C6), the levels were significantly higher at Jonsvatnet than Granåsen (t-test, $p < 0.001$). The higher levels of PFHxA in Jonsvatnet than Granåsen could potentially reflect a local source for short chained PFCAs near Jonsvatnet. In the soil and earthworm samples, there was no significant differences in PFHxA concentrations between the two study areas, while there were significantly higher concentrations of the short-chained PFBA at Jonsvatnet, compared to Granåsen. This suggests that there is not a local release of specific short-chained PFCAs to the environment near Jonsvatnet but rather a probable source of PFCA precursors, such as fluorotelomer alcohols (FTOH). Biotransformation of FTOH could explain the higher PFHxA levels in Bank voles from Jonsvatnet because this is one of the major metabolites of FTOH metabolism in

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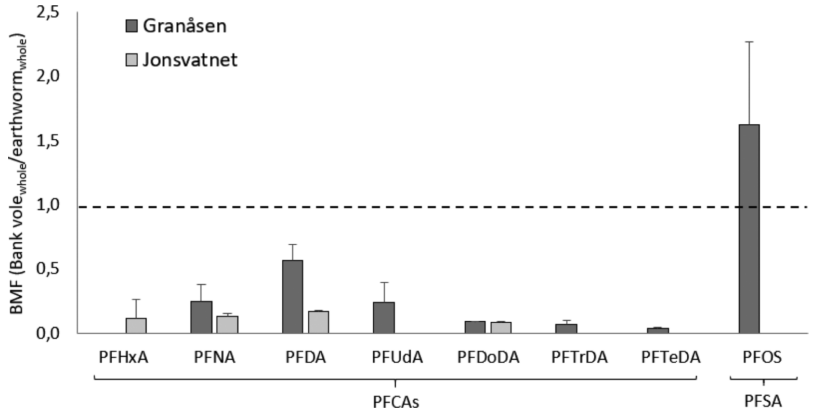

Figure 2. BMF for Bank vole$_{whole}$/earthworm$_{whole}$ (whole body concentration, w.w./w.w.) for individual PFASs in Granåsen and Jonsvatnet. Ratios are calculated from estimated (Bank vole) and measured (earthworm) average PFAS concentrations. Concentrations of PFUdA, PFTrDA, PFTeDA, and PFOS were below LOQ in the Bank voles at Jonsvatnet, and PFHxA was below LOQ at Granåsen and could not be calculated. The horizontal dashed line indicates the BMF threshold. Error bars indicate SD of the ratio (Bank vole/earthworm).

rats and other small rodents.[40] A similar observation was reported from the Antarctic Peninsula, where several PFCA compounds (e.g., PFBA, PFHxA, and PFHpA) were reported.[41] These findings suggest that the PFCAs most likely originated from FTOHs[41] because increasing trends of PFCA precursors (i.e., FTOHs) were previously observed in the Arctic with doubling times of 2.3−3.3 years between 2006 and 2012.[42]

The observed differences in PFHxA could potentially also be due to differences between locations in soil microbial communities, affecting degradation of PFCA precursors.[43−45] Furthermore, there could be differences in rate of removal between the two environments. Short-chained PFCAs readily leach from soil, and the occurrence may vary rapidly between sites, depending on the soil type. The differences in bioavailability to earthworms at the two sites or differences in the bioaccumulation pattern between earthworm and Bank vole at the two sites can probably explain these variabilities.

PFOS was the most predominant compound in Bank voles at Granåsen (Figure 1c), and the levels were higher at Granåsen, compared to Jonsvatnet, where 72% of the samples had levels below the LOQ. This is in contrast to what was measured in the soil and earthworm samples (Figure 1a,b), where PFOS concentrations were higher at Jonsvatnet, compared to Granåsen, and where PFOS was not the predominant PFAS. In wildlife studies, PFOS is usually the congener found at the highest concentrations.[46−48] However, previous studies on ski products have reported that PFCAs are the major PFASs measured in these products, while PFOS is the only PFSA detected, although at lower concentrations than the PFCAs.[5] Nevertheless, PFOS was used in skiing products in Norway until phased out in 2004. Because PFOS is very persistent, the PFOS levels measured in Bank voles in the skiing area could reflect previous use. In addition, some precursors, such as perfluorooctanesulfonyl fluoride (POSF)-based compounds[49] and perfluoroalkane sulfonamido alcohols and acrylates, degrade to PFOS.[50] However, because the same pattern was not observed in soil and earthworms, other factors such as leaching from soil, differences in the biotransformation rate, or the bioconcentration rate might be playing significant roles.

The Canadian Environmental Protection Act[51] set the liver PFOS critical toxicity value at 14.4 μg/g, based on laboratory studies in rats. Hoff et al.[38] extrapolated the environmental toxicity value for mammals to 0.144 μg/g. In the present study, the concentrations of PFOS in wild Bank voles did not exceed this value in individual animals (the maximum measured PFOS concentration was 0.016 μg/g). Accordingly, the liver concentration of PFOS detected in the Bank vole population at Granåsen may not pose a toxicological risk to these small rodents. However, considering that Bank voles are subjected to a complex mixture of PFASs, where PFOS only represents about 21%, there is still reason for concern on the physiology, endocrine, reproductive, and general health of this species and other biota at skiing areas.

While most of the research on PFASs has focused on the effects of single compounds, especially PFOS and PFOA, several hundreds of other per- and polyfluorinated compounds are currently in use[52,53] and the knowledge about the potential toxicological effects of PFAS mixtures are limited or almost nonexistent.[54] This indicates that, although their concentrations in the environment and biota are not high, they could still pose significant risks to exposed individuals under complex mixture exposure scenarios. In addition, it should be noted that the measured concentrations reported herein were detected in young individuals collected in the early summer, just after the Bank voles have started their annual reproduction cycle. The reproductive period is an exceptionally vulnerable period for these rodents, and most of the studied individuals were less than two months old, indicating that they have been exposed in utero[55] and/or from an early life stage and throughout their ontogenetic developmental period. Thus, toxicity thresholds are probably lower, compared to observations in adult rodents because young developing animals are considered more susceptible to toxic effects, compared to adults.[56]

Biomagnification of PFASs. Through the process of biomagnification, PFASs can be transferred up the food chain, where concentrations increase from one trophic level to the next via dietary accumulation.[57] The PFAS concentrations were higher in earthworms (on a w.w. basis) compared to soil (on d.w. basis) in both study locations. When considering that water in the soil will dilute the PFAS soil concentrations, the present study shows a clear bioaccumulation of PFASs from soil to earthworms. Higher concentrations in earthworms than soil were also reported in the skiing area in Oslo.[20]

The results showed that based on estimated whole-body concentrations of PFASs in the Bank voles, the BMF of PFOS at Granåsen was 1.6 (Figure 2), while for all other PFASs, the

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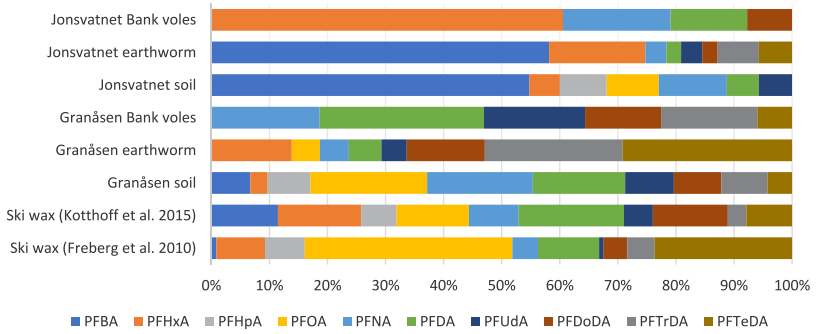

Figure 3. Overview of the contribution (%) of each congener to the total median PFCA concentration in soil, earthworm, and Bank vole samples from Gransåsen and Jonsvatnet and in ski wax samples from two different studies (Kotthoff et al. 2015[2]) and (Freberg et al. 2010[5]).

calculated BMFs were <1 in both study areas. This indicates that PFOS seems to biomagnify from earthworm to Bank vole in the skiing area, while none of the other PFASs biomagnify from earthworms to Bank vole. This is in contrast to the results from a study on a terrestrial food web (lichen-caribou-wolf),[28] which reported that several PFASs biomagnified. In that particular study, the trophic magnification factor (TMF) was the highest for PFOS and PFCAs with nine to eleven carbons. Although the BMFs for most substances except for PFOS in the present study where <1, the pattern (i.e., relative BMF) at Granåsen is comparable to the pattern found in the study on the lichen-caribou-wolf study.[28]

There may be several causes for the apparent lack of BMF of the PFASs, other than PFOS, in the present study. The Bank vole is mainly a herbivore,[58] with a diet consisting of roots, seeds, buds, and berries, in addition to earthworms and other invertebrates. On the other hand, the earthworms consume soil microorganisms, organic matter, dead leaves, and grass, and thus, the trophic levels of our study species may not be significantly different. It is therefore necessary that future studies should include organisms at a higher trophic level of the food web (e.g., carnivorous mammals or birds of prey), to properly answer whether these PFASs biomagnify in the terrestrial food chain. A recent study in the same area (county of Trøndelag) has found higher concentrations of PFASs in terrestrial birds of prey,[59] indicating that these PFASs are transported and biomagnified in terrestrial food chains.

Comparison between the PFAS Pattern. The PFCA pattern (Figure 3) is quite similar in ski wax and soil samples from Granåsen, especially for the ski wax analyzed in 2015.[2] The earthworm and Bank vole samples from Granåsen also have similar pattern to ski wax and soil samples, dominated by the longer-chained PFCAs. The long-chained PFCAs (C8−C14) make up 70−100% of the total PFCA burden in all these samples, while in the samples from Jonsvatnet, they make up only 25−40%. It is clear that the pattern measured at Granåsen is more similar to the ski wax profile than the pattern measured at Jonsvatnet (Figure 3). This strengthens the concern that ski products are a significant source of long-chained PFCAs at the local environments around skiing areas.

Studies from skiing areas found that the major PFAS congeners measured were C10−C14 PFCAs.[6] This is consistent with the findings from the present study, showing that the C10−C14 PFCAs were significantly higher at Granåsen than at Jonsvatnet. Although studies on ski products reported that PFOA is one of the main PFASs,[2,5] and it was present in the soil and earthworms at Granåsen; PFOA was not detected in the Bank voles at Granåsen. A possible reason for the low-detection frequency of PFOA in Bank voles could be the reduction of PFOA use in consumer products in Norway during the last decade, as PFOA is on Norway's priority list of chemicals, with an aim of stopping the release completely by 2020.[60] Although PFOA concentrations in soil and earthworm samples at Granåsen were below 1 ng/g, bioaccumulation of PFOA should be expected in the voles.[61] Thus, it is surprising that PFOA was not found in the Bank vole samples.

In summary, the different PFAS pattern in the two study areas clearly shows that there are different sources of PFASs to these two environments. However, the detected concentrations are far below toxicity threshold levels set in laboratory studies, indicating that individual PFASs in ski products may not pose a significant risk to the environment. Still, it should be taken into consideration that the reported concentrations were measured in organisms from the base of the food web, and because PFASs are persistent, and several of the PFASs biomagnify in food webs,[28] the levels could be much higher at a higher trophic level, such as top predators. In addition, they are exposed to a mixture of PFASs, rather than single contaminants, so the issue of mixture toxicity should also be considered and addressed in any risk environmental assessment program of contaminants from skiing areas.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b02533.

Map of the study areas, internal standards used for quantification of PFASs, PFASs in soil samples from Jonsvatnet and Granåsen, PFASs in earthworm samples from Jonsvatnet and Granåsen, PFASs in Bank vole samples from Jonsvatnet and Granåsen, PFASs in blank samples, additional analytical details of sampling, chemical analyses, and data treatment (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: randi.gronnestad@ntnu.no. Phone: +47 924 804 75.

ORCID

Randi Grønnestad: 0000-0002-2968-4077

Augustine Arukwe: 0000-0002-2134-4799

Bjørn Munro Jenssen: 0000-0002-7042-2191

Author Contributions

The manuscript was written through contributions of all the authors. All the authors have given approval to the final version of the manuscript.

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Notes

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■ ABBREVIATIONS

LOD	limit of detection
LOQ	limit of quantification
BMF	biomagnification factor
TMF	trophic magnification factor
PFAS	perfluoroalkyl substances
PFCA	perfluorocarboxylic acid
PFSA	perfluorosulfonic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctansulfonate
POSF	perfluorooctanesulfonyl fluoride
PFBA	perfluorobutanoic acid
PFHxA	perfluorohexanoic acid
PFHpA	perfluoroheptanoic acid
PFNA	perfluorononanoic acid
PFDA	perfluorodecanoic acid
PFUdA	perfluoroundecanoic acid
PFDoDA	perfluorododecanoic acid
PFTrDA	perfluorotridecanoic acid
PFTeDA	perfluorotetradecanoic
PFBS	perfluorobutane sulfonate
PFHxS	perfluorohexane sulfonate
FASAs	perfluoroalkane sulfonamide derivatives
FOSA	perfluoro-1-octane sulphonamide
N-MeFOSA	*N*-methyl perfluoro-1-octane sulphonamide
N-EtFOSA	*N*-ethyl perfluoro-1-octane sulfonamide
N-MeFOSE	2-(*N*-methyl perfluoro-1-octane sulfonamido) ethanol
N-EtFOSE	2-(*N*-ethyl perfluoro-1-octane sulfonamido) ethanol
w.w.	wet weight
d.w.	dry weight

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