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Turfgrass Environmental Impacts

Leaching and surface runoff after fall application of fungicides on putting greens

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Abstract

Many greenkeepers and authorities are concerned about the environmental risks resulting from pesticide use on golf courses. We studied leaching and surface runoff of fungicides and metabolites for two winter seasons after fall application of boscalid, pyraclostrobin, prothioconazole, trifloxystrobin, and fludioxonil in field lysimeters at NIBIO Landvik, Norway. The applications were made on creeping bentgrass greens (5% slope) that had been established from seed or sod (26-mm mat) on USGA-specification. root zones amended with Sphagnum peat or garden compost, both with 0.3–0.4% organic carbon in the root zone. The proportions of the winter precipitation recovered as surface and drainage water varied from 3 and 91% in 2016–2017 to 33 and 55% in 2017–2018 due to differences in soil freezing, rainfall intensity, and snow and ice cover. Detections of fungicides and their metabolites in drainage water were mostly within the environmental risk limits (ERLs) for aquatic organisms. In contrast, concentrations in surface runoff exceeded ERLs by up to 1,000 times. Greens established from sod usually had higher fungicide losses in surface runoff but lower losses in drainage water than greens established from seed. Presumably because of higher microbial activity and a higher pH that made prothioconazole-desthio more polar, fungicide and metabolite losses in drainage water were usually higher from greens containing compost that from greens containing peat. Leaching of fungicides and metabolites occurred even from frozen greens. The results are discussed in a practical context aiming for reduced environmental risks from spraying fungicides against turfgrass winter diseases.

1 | INTRODUCTION

Microdochium patch caused by *Microdochium nivale* (Fr.) Samuels & Hallett affects turfgrasses both in the fall and under snow cover during winter and is the economically most important disease on Nordic golf courses. Golf courses in areas with at least 1 mo of snow cover are also affected by gray snow mold caused by *Typhula incarnata* Fr., and areas with at least 3 mo of snow cover are affected by speckled snow mold caused by *Typhula ishikariensis* Imai (Årsvoll, 1975). A survey in 2014–2015 showed that most Nordic greenkeepers treat their greens with fungicides once or twice in the fall against these diseases (Økland et al., 2018). Within each of

Abbreviations: DT50, Dissipation half-life; ERL, Environmental risk limit; Koc, Sorption coefficient of organic carbon the Nordic countries, three to six active fungicide ingredients are currently approved for control of microdochium patch and snow molds (Espevig & Aamlid, 2018).

One of the greatest concerns with fungicide use on golf courses is that the active substances or their metabolites find their way to streams, rivers, lakes, and ground water (Baris, Cohen, Barnes, Lam, & Ma, 2010; Beard & Kenna, 2008). The EU has a general safety limit of 0.1 μg L^{-1} of any pesticide in drinking water, and Sweden and Norway have established environmental risk limits (ERLs) for individual pesticides and their metabolites for protection of aquatic organisms (Anderson & Kreuger, 2011; Norwegian ERL Database, 2019; Stenrød, 2015).

Former Scandinavian research on fungicide leaching from sand-based putting greens (reviewed by Aamlid, 2014) found that (a) the risk for leaching to ground and surface water depended on the chemical properties of the fungicides, particularly their sorption coefficients and half-lives; and (b) that the risk for fungicide leaching could be almost eliminated by increasing the organic matter content in the sand-based root zone (0–30 cm; USGA, 2018) from less than 1 to 2–3% (w/w; Larsbo, Aamlid, Persson, & Jarvis, 2008; Strömqvist & Jarvis, 2005; Aamlid et al., 2009). However, those studies were mostly conducted with fungicide that are not on the market anymore, and they did not include metabolites, that is, products that an active fungicide ingredient is broken down to and that may often be equally or more harmful to the environment than the active ingredient itself. While focusing on organic matter content, these studies also paid little attention to *type* of organic matter in the root zone. Up to now, the organic amendment most commonly used in sand-based greens has been *Sphagnum* peat. However, in Norway, the government has proposed a ban on the use of peat in horticulture starting in 2030 because of the CO_2–emissions resulting from the excavation and processing of peat from bogs (Boldrin, Hartling, Laugen, & Christensen, 2010). For sand-based putting greens, the most likely alternative are different types of composts (Favoino & Hogg, 2008); however, root zones amended with compost typically have much higher pH values than substrates amended with *Sphagnum* peat (Aamlid, 2005; Mandelbaum & Hagar, 1990), and this may well affect the sorption and risk for leaching of certain fungicides (Wauchope et al., 2002). Root zones amended with compost may also have higher microbial activity (Mandelbaum & Hagar, 1990; Aamlid et al., 2009; Niklasch & Jørgensen, 2001), perhaps leading to faster fungicide degradation than in root zones amended with peat.

A characteristic feature of turfgrass areas, especially sand-based putting greens, is the thatch, an intermingled organic layer of dead and living shoots, stems, and roots that develops between the turf canopy and the soil surface (Beard, 2002). Upon topdressing, the thatch layer is gradually diluted into a mat layer. The thatch or mat layer is likely to have a strong impact on the risk for pesticide leaching as is represents a significant barrier to the penetration of pesticides into the soil as well as a potential site for fungicide accumulation (Carroll, 2008; Cisar & Snyder, 1996). This role of thatch was not addressed in former Scandinavian projects, and U.S. reports have shown variable results as to the efficacy of thatch in reducing pesticide leaching (Sigler, Taylor, Throssell, Bischoff, & Turco, 2000). Models developed for the sorption of non-ionic organic compounds to soil organic matter tend to overestimate their sorption to thatch organic matter (Lickfeldt & Branham, 1995). In some cases, it has even been shown that the thatch layer prevents pesticides from getting into contact with the underlying root zone, thus decreasing fungicide degradation and increasing the risk for surface runoff (Sigler et al., 2000). This problem may also occur when establishing new turfgrass areas from sod, as Canaway (1993) observed a much stronger reduction in infiltration rates for sodded than for seeded football pitches during the first year after establishment. The establishment and repair of putting greens with sod is common on Scandinavian golf courses, but the impact of this practice on fungicide behavior has not been investigated previously.

Core Ideas

- Surface runoff had fungicide concentrations many times higher than ERLs.
- Metabolites of prothioconazole, trifloxystrobin, and fludioxonil were found in drain discharge.
- Leaching occurred even in periods with frozen greens.
- Sodded greens had higher surface losses, but lower leaching losses, than seeded greens.
- Leaching of metabolites were higher from sand amended with compost instead of *Sphagnum* peat.

Putting greens are often undulated to make them more challenging to the players, and although infiltration rates are usually higher than on natural soils, it is commonly observed that water moves on the surface to the lower parts on the green. From environmental monitoring of turfgrass areas in United States, there are several examples of alarming pesticide concentrations in surface water, especially during the first storm water event after pesticide application (e.g., King & Balogh, 2013; Kramer et al., 2009; Petrovic & Easton, 2005; Slavens & Petrovic, 2012). Since those studies were conducted during the growing season with turfgrass growing on natural soils, there is, however, a need for more knowledge about the risk for fungicide runoff from sand-based greens when fungicides are sprayed in the late fall for control of winter-active diseases. The need for such studies is further underlined by the

Intergovernmental Panel on Climate Change's predictions for a milder and more unstable winter climate with higher and more intensive precipitation in northern areas (IPCC, 2019).

Pesticide runoff in arable agriculture is commonly correlated with soil erosion, and the estimated losses per unit area are usually a function of plot size (Southwick, Meek, Fouss, & Willis, 2000). In contrast, Carroll, Hapeman, and Pfeil (2009) found that there was no problem in upscaling data from small fairway plots to account for what would occur at the edge of a golf course fairway, and this will most likely be correct even putting greens with a 100% cover of uniform, high-density, and short-cut turf. The risk for pesticide losses in surface runoff are related to pesticide properties like the sorption coefficient, water solubility, degradation half-lives, dose, and application rate, in addition to soil type and climate (Rice, Horgan, & Rittenhouse, 2010). The sorption coefficient (Koc) of the pesticide may vary with organic matter decomposition and are not necessarily the same for sand-based root zones as for agricultural soils (Carroll & Leshin, 2010).

The objectives of the research reported here were to (a) clarify the risk for leaching and surface runoff of fungicides currently approved or considered for approval for control of turfgrass winter diseases in the Nordic countries, including important metabolites; (b) compare fungicide leaching and surface runoff from putting greens with Sphagnum peat vs. garden compost as organic amendment to the sand-based root zone; and (c) determine the effect of a mat layer high in organic matter on the risk for fungicide losses to the environment.

2 | MATERIALS AND METHODS

2.1 | Experimental site

A field experiment was conducted in the lysimeter facility at the NIBIO Turfgrass Research Center Landvik, Norway (58˚19′N; 8˚30′E, 5 m asl) from 25 Oct. 2016 to 20 Mar. 2017 and from 18 Oct. 2017 to 6 Apr. 2018. The facility consisted of 16 stainless steel lysimeters arranged in four blocks. Each lysimeter was 2 m long, 1 m wide and placed in the center of a 3 m × 2 m plot to avoid border effects. Each lysimeter was filled with a 30- to 40-cm layer of sand above a 10- to 15-cm gravel layer according to the recommendations from United States Golf Association (USGA, 2018). The gravel was placed directly on the sloping bottom of the lysimeters which directed water to the lysimeter outlet and further to a 200-L stainless steel container for collection of drainage water. In preparation for this experiment, the lysimeters had been deturfed, more root zone material added, and the surface reshaped to a slope of 5%. This allowed collection of surface runoff from the 2 m^2-lysimeter surface through a 1-m wide trench leading to a 25-L stainless steel container.

2.2 | Experimental treatments and design

The experiment had two factors, each with two levels. In Factor 1, we compared fungicide leaching and runoff from USGA specification. root zones amended with *Sphagnum* peat vs. Garden compost (hereafter referred to as 'Peat' and 'Compost', respectively) and in Factor 2, we compared fungicide leaching and runoff from newly seeded greens (no thatch) vs. greens established using sod with a 26-mm mat layer.

In Factor 1, our intention was to compose substrates with a similar content, but different origin and quality of organic matter, thus resulting in different potential for fungicide degradation. The pH of the substrates was measured in a water/soil ratio (v/v) of 2.5 by use of a Radiometer PHM210 pH meter and a Thermo Ross pH electrode after staying overnight at room temperature. Plant available P, K, Mg, Ca, and Na was determined after extraction with a solution 0.1 M ammonium-lactate and 0.4 M acetic acid (Egnér, Riehm, & Domingo, 1960). Total C and total N were determined using a LECO TrueSpec analyzer where C was measured as CO_2 by an infrared cell, and N was measured as N_2 by thermal conductivity after digestion of the sample at 1,050 ˚C (Nelson & Sommers, 1996). Total C was taken as organic C when the pH was 6.5 or less and the C/N ratio calculated as the ratio between total C and total N. The cation exchange capacity was measured after extraction with 1 M ammonium acetate at pH 7.00. Three g of soil were washed with small portions of the extraction solution to 250 ml in a volumetric flask and Ca, Mg, K, and Na measured in the extract by use of ICP-OES. Determination of H^+ concentration was done by titrating the percolate back to pH 7.00 using NaOH.

The soil chemical analyses of the two substrates showed similar cation exchange capacity and concentrations of organic C and plant-available K and Mg, but a lower C/N ratio, higher pH, and concentration of plant-available P in the compost-amended root zone (Table 1).

The turfgrass used on both seeded and sodded plots was established from a creeping bentgrass (*Agrostis stolonifera* L.) seed blend containing 'Penn A4', 'Penn G2', and 'Penn G6' (33.3% [w/w] of each variety). The 30-mo-old sod had been grown on USDA-specification. sand and had a thickness of 26 mm. Seeding and sodding took place in May 2016. Soil samples taken from the 0- to 3-cm top layer before the first fungicide application in October 2016 showed identical pH but the cation exchange capacity and concentrations of organic C and plant available P, K, and Mg were 51, 164, 77, 79, and 93% higher for sodded than for seeded turf, respectively (Table 1).

The experimental green was maintained according to good greenkeeping practice, including mowing with a walk-behind single mower to 3 mm three times per week and light topdressing once a week for a total height of 5.9-mm sand in 2016 and

TABLE 1 Soil chemical propertied as affected by organic amendment to the USGA-specification sand (USGA, 2018) used for construction and establishment method. Samples were before the first fungicide application in October 2016 and sieved thoroughly before analysis

	Organic C	C/N	P-AL[a]	K-AL[a]	Mg-AL[a]	pH[b]	CEC[c]
	kg (kg soil)$^{-1}$		mg (kg soil)$^{-1}$				cmol c+ (kg soil)$^{-1}$
Organic amendment[d]							
Peat	0.38	45	13	28	17	5.5	6.8
Compost	0.29	20	28	24	15	6.5	6.7
Establishment method[e]							
Seeded green	0.28	16	48	126	27	5.8	6.1
Sodded green	0.62	22	85	225	52	5.8	9.2

[a]AL extraction: 0.1 M ammonium-lactate + 0.4 M acetic acid.
[b]pH measured in distilled water.
[c]CEC, cation exchange capacity.
[d]Soil samples taken at 3- to 30-cm depth.
[e]Soil samples taken at 0- to 3-cm depth.

6.6-mm sand in 2017. Verticutting was performed four times on seeded plots and six times on sodded plots in 2016. In 2017, all plots were aerated 10 times to 15-mm depth using a slicer with knives 40-mm apart. In 2017, the plots were also subjected to wear from a friction wear drum with golf spikes corresponding to 15,000 rounds of golf. Fertilizers, partly liquid (Wallco 5–1–4 NPK, Orkla Care, Solna, Sweden) and partly granular (Greenmaster Cold Start 11–2.2–4.1 NPK in spring and Greenmaster 14–08.3 NPK in summer and fall; ICL Specialty Fertilizers, Ipswich, UK) were applied every 2 wk for a total rate of 26 g N m^{-2} on seeded plots and 18 g N m^{-2} on sodded plots in the grow-in year 2016 and 15 g N m^{-2} on all plots in 2017.

The fungicides prothioconazole (*RS*)−2-[2-(1-chlorocyclopropyl)−3-(2-chlorophenyl)−2-hydroxypropyl]−2,4-dihydro-1,2,4-triazole-3-thione), trifloxystrobin (methyl (*E*)-methoxyimino-{(*E*)-α-[1-(α,α,α-trifluoro-m-tolyl)ethylideneaminooxy]-o-tolyl}acetate), boscalid (2-chloro-*N*-(4′-chlorobiphenyl-2-yl)nicotinamide) and pyraclostrobin (methyl {2-[1-(4-chlorophenyl)pyrazol-3-yloxymethyl]phenyl} (methoxy) carbamate) were applied on all plots on 25 Oct. 2016 and 18 Oct. 2017 at the following rates: 175, 150, 400, and 100 g a.i. ha^{-1}, respectively. Prothioconazole and trifloxystrobin were applied as the commercial product Delaro SC 325 (Bayer Crop Science, Leverkusen, Germany; 175 and 150 g a.i. L^{-1}, respectively), and boscalid and pyraclostrobin were applied as the commercial product Signum (BASF, Ludwigshafen, Germany; 267 and 67 g a.i. kg^{-1}, respectively). The two commercial products were not tank-mixed but sprayed separately 2 h apart. After mowing had been discontinued for the season, fludioxonil (4-(2,2-difluoro-1,3-benzodioxol-4-yl)−1*H*-pyrrole-3-carbonitrile),) was applied on all plots at a rate of 375 g a.i. ha^{-1} on 15 Nov. 2016 and 8 Nov. 2017. Fludioxonil was applied as the commercial product Medallion TL (125 g a.i. L^{-1}; Syngenta, Basel, Switzerland). The fungicides were applied in a water volume of 250 L ha^{-1} using a modified Oxford experimental backpack plot sprayer (Norsprayer, Gabrielsen Maskinforretning, Kristiansand, Norway) working at 150–200 kPa pressure. The actual application rates were recorded by weighing the tank before and after spraying to ensure that deviations from target rates were less than ± 10%.

Table 2 shows that the dissipation half-life (DT50) rates of the studied fungicides are quite different from each other. Degradation of prothioconazole, trifloxystrobin, and pyraclostrobin was expected to be fast due to aqueous photolysis after spraying, whereas boscalid was expected to be rather stable. We included the most important metabolites or transformation products of the fungicides in the water analysis in order to better assess leaching and runoff patterns. The sorption coefficients in (agricultural) soils are also widely different for the compounds in the study, as well as their toxicity to water dwelling organisms.

2.3 | Weather data

In both years, the experimental period was milder and had more precipitation than the 30-yr (1961–1990) reference period (Table 3; Figure 1). The second winter (2017–2018) was colder and implied a longer duration of frozen greens and snow cover, but also more fluctuations between cold and mild weather resulting in ice formation and more surface runoff than in the first year. In the first year, there was a record-high precipitation of 147 mm d^{-1}, starting as rain and turning into snow on 5 November, 10 d after the first fungicide application (Figure 1a). In the second year, there was also a week with rather high precipitation values after the first fungicide application on 18 October (Figure 1b).

TABLE 2 Some properties of the studied pesticides and metabolites, data from Pesticide Properties DataBase (PPDB, 2020) and Norwegian ERL Database (2019). ERL, Norwegian environmental risk limit for water dwelling organisms

Pesticide or metabolite	DT50, Aqueous photolysis[a]	DT50, Field soil[b]	Sorption Kfoc[c]	ERL[d]
	d		L kg^{-1}	µg L^{-1}
Boscalid	Stable	254	772	12.5
Pyraclostrobin	0.06	33	9,315	0.40
Pyraclostrobin metabolite BF 500-6	n.a.[e]	506	60,495	n.a.
Prothioconazole	2.1	0.8	2,556	0.74
Prothioconazole-desthio	55	25	575	0.033
Trifloxystrobin	2.7	1.7	2,287	0.192
Trifloxystrobin acid	1.7	70	116	320
Fludioxonil	10	21	132,100	0.050
Fludioxonil metabolite CGA 192155	n.a.	19	24	100

[a]Dissipation half-life in water exposed to sunlight.
[b]Dissipation half-life in field soil.
[c]Freundlich sorption coefficient to soil organic carbon (PPDB, 2020).
[d]ERL, Norwegian environmental risk limits (Norwegian ERL Database, 2019).
[e]n.a., not available.

TABLE 3 Mean monthly temperature, total precipitation, days with frozen soil and days with snow and/or ice cover during the experimental periods in 2016-2017 and 2017–2018. Monthly temperatures and precipitation data have been compared with the 30 yr reference period 1961–1990

	Temperature mean			Precipitation		
	°C			mm		
	2016–2017	2017–2018	30-yr normal	2016–2017	2017–2018	30-yr normal
Oct. (after start of trial)	7.4	8.5	–	4	237	–
Nov.	2.7	3.5	3.2	256	157	143
Dec.	3.7	1.7	0.2	44	116	102
Jan.	1.7	0.9	−1.6	65	222	113
Feb.	0.4	−2.0	−1.9	139	143	73
Mar. (whole month)	3.4	−1.2	1.0	118	49	85
Apr. (until end of trial)	–	2.0	–	–	25	–
Mean or sum, Nov.–Mar.	2.4	0.6	0.2	621	686	516
Mean or sum, trial period	2.3	1.3	–	601	948	–
Days with frozen soil	82	127				
Days with snow or ice cover	30	78				

2.4 | Data collection

2.4.1 | Sorption studies

The sorption coefficients Kd (sorption to soil) and Koc (sorption to soil organic carbon) for boscalid; pyraclostrobin, the prothioconazole metabolite prothioconazole-desthio (α-(1-chlorocyclopropyl)-α-o(2-chlorophenyl)methyl-1*H*-1,2,4-triazole-1-ethanol); and fludioxonil were determined at two soil depths (0–3 and 3–30 cm) in accordance with the Guideline 106 (OECD, 2000). The soils were sampled from the greens in September 2016. Boscalid and pyraclostrobin were added as the commercial product Signum and fludioxonil was added as the commercial product Medallion. Prothioconazole-desthio was added as the pure compound (pestanal purity standard Supelco, Sigma Aldrich, Darmstadt, Germany). A soil/solution ratio of 1:10 was used for boscalid, pyraclostrobin, and prothioconazole-desthio, whereas a ratio of 1:25 was used for fludioxonil. Prothioconazole and trifloxystrobin were not included in the sorption study due to the instability of the parent compounds, and the metabolites trifloxystrobin acid, BF 500-6 (metabolite of pyraclostrobin), and CGA 192155 (2,2-difluoro-benzo(1,3)dioxol-4-carbocyclic acid; metabolite of fludioxonil) were not included because toxicities were very low or had not been determined. After removal of plant material, 1- or 2.5-g portions of air dried, sieved (2-mm

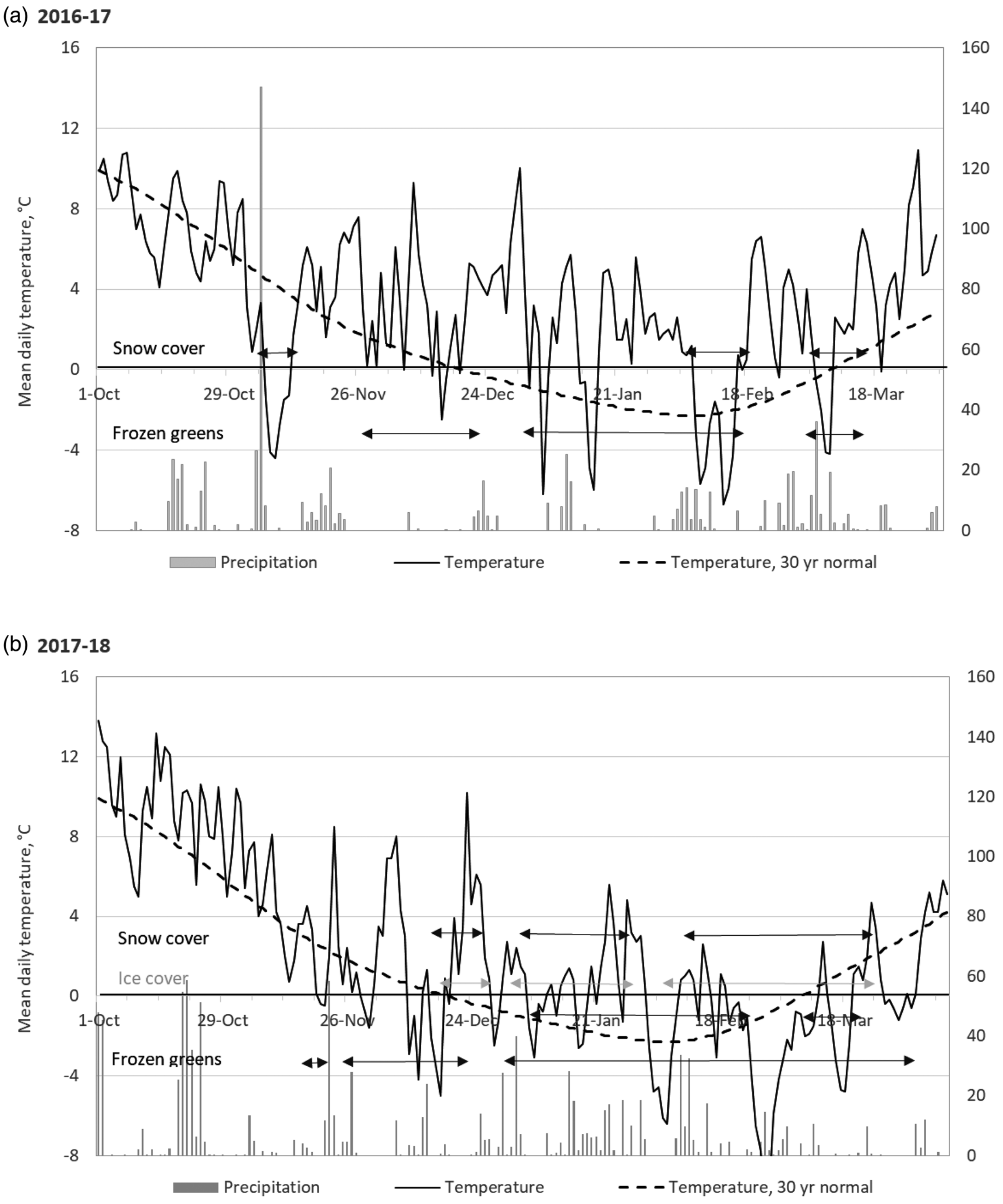

FIGURE 1 Mean daily temperature (compared with the 30-yr normal value, 1961–1990), daily precipitation, and days with frozen greens and snow and ice cover during the experimental periods in (a) 2016–2017 and (b) 2017–2018 (There was no ice cover in 2016–2017)

mesh) soil was weighed in triplicate into Teflon centrifuge tubes (40 ml) and mixed with 23 ml 0.01 M $CaCl_2$. After 16 h of pre-equilibration on a horizontal shaker at room temperature (20 ˚C), the pesticide solution was added to the suspensions to give final concentrations in the range of 1.4 to 5.6 µg (g soil)$^{-1}$. After further equilibration by shaking for 24 h, the suspensions were centrifuged for 10 min at 9220 × g. The supernatant (750 µl) was transferred to a LC-vial containing 250 µl methanol and internal standards of metconazole and 2,4-D and mixed before immediate LC-MS/MS determination of pesticide content followed by calculation of the sorption coefficients (Kd and Koc).

2.4.2 | Soil porosity, hydraulic conductivity, infiltration, and frost depth

One undisturbed soil sample, 37-mm high and 58-mm diameter, was taken from each of the depths 5–42 and 150–187 mm just outside the lysimeter in each plot (not to disturb hydraulic properties within the lysimeter) shortly before the first fungicide application in October 2016. The samples were analyzed for bulk density and air-filled and water-filled porosity at a pressure potential of −2.45 kPa. Air permeability was determined according to Green and Fordham (1975), and saturated hydraulic conductivity was estimated from air permeability according to Riley (1996). On the same day and at the start of the second experimental period in October 2017, turfgrass infiltration rates were measured using a double ring infiltrometer with 120- and 50-mm diameter of the outer and inner ring, respectively. The infiltrometer was filled to a height of 80 mm and infiltration measured after 3 min at two random sites per plot.

In the second experimental year, frost tubes, 8-mm diameter, 40-cm deep, and filled with methylene-blue (Iwataa, Horota, Suzuki, & Kuwao, 2012), were installed at the top and bottom of the slope on each plot in two out of four blocks in the lysimeter facility. The depth of the upper and lower border between frozen and thawed root zone was measured two to three times per week throughout the winter.

2.4.3 | Collection of drainage and surface runoff and LC-MS/MS analysis

Two d after each fungicide application, the amounts of drainage and surface water were measured, collectors emptied, and the first water samples taken for analyses of the fungicides and their metabolites. Later samplings and volume measurements of drainage and surface water were performed every time the collectors were full. Because of more precipitation and longer periods with frozen soil, 24 samples of surface water were taken from each plot in 2017–2018 as opposed to only seven samples in 2016–2017. For drainage water, the corresponding number of samples per plot was 10 in 2016–2017 and 12 in 2017–2018.

The water samples were stored frozen until pesticide analysis. The surface water samples had so high fungicide concentrations that they could be analyzed directly by LC-MS/MS without pre-concentration. These samples were filtered (Phenomenex regenerated cellulose, 0.45 µm), and 950 µl were transferred into vials containing 50 µl internal standard solution (metconazole, pyraclostrobin-d3 and 2,4-D) and analyzed. The drainage water samples were pre-concentrated by solid-phase extraction after removal of particles by Büchner filtration (Sartorius filter paper No. 393, 90 mm, 100 g m^{-2}). For each sample, 200 ml (containing internal standard mix) was passed through a pre-conditioned Strata X-AW sorbent (200-mg sorbent mass). The sorbents were dried by airflow and the fungicides and metabolites eluted from the sorbent using 4 ml of 5% (w/w) formic acid in acetone. The eluate was reduced to dryness under a flow of nitrogen gas, re-dissolved in 0.5 ml of methanol, and filtered (Phenomenex RC 0.45 µm, 4-mm syringe filter) into an LC-vial. Thus, each 200-ml drainage water sample was pre-concentrated into 0.5 ml.

The fungicide and metabolite concentrations were measured on a Waters Alliance 2695 LC-system coupled to a Quattro Ultima Pt triple quadropole mass spectrometer (Micromass, Manchester, UK). A 5-µl sample volume was injected, and the analytes separated on a Phenomenex Kinetex Biphenyl column (100 by 2.1 mm, particle diameter 5 µm) using 5 mM formic acid and methanol as mobile phases. The mass spectrometer was used in electrospray polarity switching mode to detect all analytes within a runtime of 15 min. Concentrations were calculated using five-point internal standard calibration at 1 to 200 ng ml^{-1}, with reference standards of boscalid, pyraclostrobin, pyraclostrobin metabolite BF 500-6, trifloxystrobin, trifloxystrobin acid metabolite, prothioconazole, prothioconazole-desthio, fludioxonil, and fludioxonil metabolite CGA 192155, all from Dr. Ehrenstorfer GmbH, Germany, except the metabolite BF 500-6 which was a kind donation from Bayer Crop Science, Leverkusen, Germany. The internal standards (pyraclostrobin-*N*-methoxy-d3, metconazole, and 2,4-D) were purchased from Dr. Ehrenstorfer GmbH, Augsburg, Germany. Internal standard calibration was used, at a level of 40 ng ml^{-1} in both samples and standards to adjust for any matrix effects and variability during analysis. The limit of quantification was 1 ng ml^{-1} for all compounds, except prothioconazole, fludioxonil, and CGA 192155 for which it was 5 ng ml^{-1}. This corresponds to 2.5–12.5 ng L^{-1} in drainage water. Blank and spiked control samples in MilliQ water were prepared with each batch to check for contamination and calculate analyte recoveries. Recovery was 100% in surface water and between 76 and 108% for most of the analytes in drainage water, except trifloxystrobin which had a recovery of 45%, and prothioconazole and prothiconazole-desthio which had recoveries of 14 and 131%, respectively, in drainage water. Prothioconazole is an unstable compound prone to rapid aqueous photolysis which forms the stable metabolite prothioconazole-desthio by the loss of sulfur.

2.5 | Environmental risks and statistical analyses

The calculated values for fungicide and metabolite Koc were compared with the ranges for Freundlich sorption coefficients

TABLE 4 Combined effect of seeding vs. sodding and type of organic amendment on the sorption coefficient Kd at 0- to 3- and 3- to 30-cm soil depth of the fungicides boscalid, pyraclostrobin, fludioxonil, and the prothioconazole metabolite prothioconazole-desthio. For each of the two soil layers, the mean sorption coefficients to soil organic carbon (Koc, L kg^{-1}) have also been calculated and compared with the ranges for Freundlich sorption coefficients (Kfoc, L kg^{-1}) reported in the Pesticide Properties DataBase (PPDB, 2020)

			K_d sorption coefficient ± SE (n = 2)			
Treatment	**Soil depth**	**Total organic C**	**Boscalid**	**Pyraclostrobin**	**Prothioconazole-desthio**	**Fludioxonil**
	cm	kg (kg soil)$^{-1}$	L kg^{-1}			
Seeded over peat	0–3	0.26	2.8 ± 0.18	15 ± 0.09	3.1 ± 0.42	3.3 ± 0.47
Sodded over peat	0–3	0.47	7.8 ± 0.14	49 ± 1.1	7.0 ± 0.38	24 ± 0.78
Seeded over compost	0–3	0.29	4.7 ± 0.24	23 ± 0.38	4.0 ± 0.29	6.9 ± 0.85
Sodded over compost	0–3	0.77	9.3 ± 0.57	49 ± 2.8	8.8 ± 0.78	23 ± 1.91
Seeded over peat	3–30	0.38	4.6 ± 0.21	23 ± 1.8	8.1 ± 0.50	9.8 ± 1.2
Sodded over peat	3–30	0.37	4.5 ± 0.41	24 ± 1.0	8.0 ± 0.44	8.0 ± 0.71
Seeded over compost	3–30	0.29	4.7 ± 0.24	23 ± 0.38	4.0 ± 0.29	6.9 ± 0.85
Sodded over compost	3–30	0.29	3.8 ± 0.42	23 ± 0.11	4.0 ± 0.35	8.4 ± 1.1
Koc, average for the 0- to 3-cm layer ± SE (n = 8)			1,391 ± 104	7,622 ± 735	1,301 ± 57	2,935 ± 570
Koc, average for the 3- to 30-cm layer ± SE (n = 8)			1,339 ± 68	7,100 ± 345	1,763 ± 157	2,504 ± 110
Kfoc range from PPDB (2020)			594–1,110	4,240–12,000	523–625	7,500–210,000

reported in the Pesticide Property DataBase (PPDB, 2020). The Kfoc ranges have been iterated from sorption studies using varying pesticide concentrations and various soil and were therefore considered a more correct reference point than individual Koc values reported in the literature.

The concentration of fungicides and their metabolites in drainage water and surface runoff were compared with the Norwegian ERLs (Norwegian ERL Database, 2019). These values indicate threshold concentrations above which long-term negative effects in aquatic environments might occur. The concentration limits are based on 'No Observed Effects Concentrations (NOEC)'-data from chronic toxicity tests of aquatic organisms, and the calculation includes an assessment factor depending on the quality of these data. The calculation procedure is in accordance with guidelines for environmental quality standards (EQS) for EU's Water Framework Directive (European Union, 2013).

The data were analysed using the SAS procedure PROC ANOVA (SAS Institute, Cary, NC). In the text, the term 'significant' always refers to $P \leq .05$, whereas effect with $.05 < P \leq .10$ are referred to as 'tendencies' or 'trends'. Significant differences among treatment combinations were identified using Fisher's least significant difference (LSD) at $P \leq .05$.

3 | RESULTS

3.1 | Sorption studies

The sorption studies showed four to five times stronger sorption coefficients for pyraclostrobin than for boscalid, whereas fludioxonil and prothioconazole-desthio were intermediate (Table 4). Sorption in the 3-cm top layer was always stronger on plots established by sodding than on plots established by seeding. The sod was 26-mm thick, so the sorption coefficients in the sodded 0- to 3-cm layer were expected to be mostly unaffected by the underlying root zone, which was also the case. As expected, there was also no effect of the seeded vs. sodded top layer on the sorption of any of the fungicides at 3- to 30-cm depth, that is, below the sod. Boscalid, pyraclostrobin, and fludioxonil sorbed equally well to sand amended with peat as to sand amended with compost, but prothioconazole-desthio sorbed stronger after amendment with peat. Prothioconazole-desthio is a weak acid (pKa 6.9) which will be neutral below pH 6 and therefore sorbed stronger to sand amended with peat (pH 5.5) than to sand amended with compost (pH 6.5).

After correction for organic carbon, the Koc sorption coefficients were on the same level in the 0- to 3- and 3- to 30-cm soil layers (Table 4), demonstrating that sorption was strongly related to the organic carbon content in the soils. As compared with Koc values reported for agricultural soils (PPDB, 2020), the values were higher for boscalid, pyraclostrobin, and prothioconazole-desthio, but lower for fludioxonil.

According to the International SSLRC Mobility Classification System (used by PPDB, 2020), boscalid, prothioconazole-desthio, and fludioxonil, all with Koc-values in the range of 1,000 to 4,000, could be classified as slightly mobile in the sand-based root zones, whereas pyraclostrobin, with a Koc > 4,000, could be classified as non-mobile.

TABLE 5 Main effect of experimental factors on air-filled, water-filled and total porosity at −2.45 kPa, bulk density, and saturated hydraulic conductivity (K_{sat}) as determined in undisturbed cylinder samples taken from two depths in October 2016, and on infiltration measured with a double-ring infiltrometer in October 2016 and October 2017

	5- to 42-mm depth					150- to 187-mm depth					Infiltration	
	Porosity					Porosity						
	Air filled	Water filled	Total	Bulk density	K_{sat}	Air filled	Water filled	Total	Bulk density	K_{sat}	Oct. 2016	Oct. 2017
	%			kg dm^{-3}	mm h^{-1}	%			kg dm^{-3}	mm h^{-1}	mm h^{-1}	
Peat	31.3	24.7	56.0	1.24	333	36.6	15.0	51.6	1.30	336	930	596
Compost	29.7	23.8	53.5	1.26	323	33.7	13.8	47.5	1.35	317	884	606
Significance	ns‡	ns	ns	ns	ns	***	ns	***	***	ns	ns	ns
Seed	28.9	19.3	48.2	1.44	321	34.4	14.6	48.9	1.31	304	1,123	826
Sod	31.1	29.2	61.3	1.07	336	35.9	14.2	50.1	1.34	348	691	376
Significance	*	***	***	***	ns	ns	ns	*	ns	**	**	***
Interaction	ns	ns	ns	ns	**	*	ns	*	*	*	ns	ns

*.01 < P ≤ .05; **.001 < P ≤ .01; ***P ≤ .001; ‡ns, not significant (P > .1).

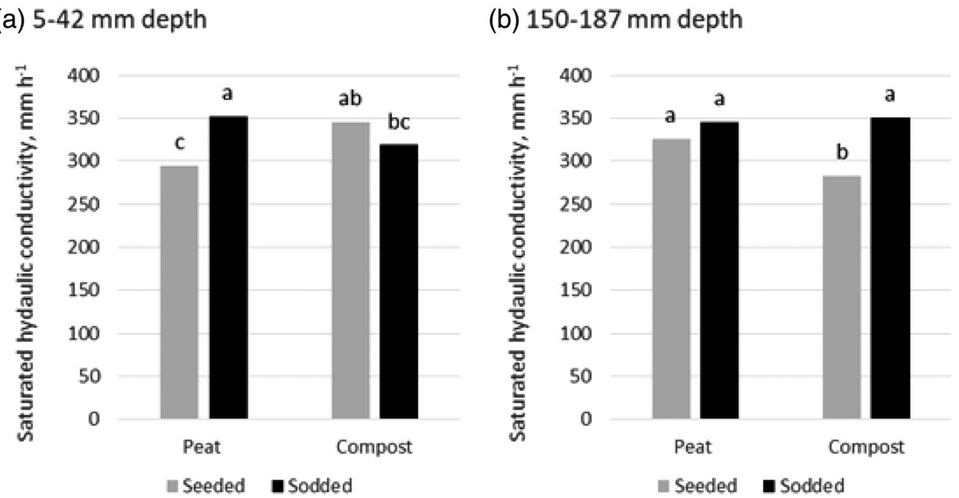

FIGURE 2 Effect of combinations of organic amendment to the sand-based root zone and turfgrass establishment method on saturated hydraulic conductivity in cylinder samples taken at 5- to 42- and 150- to 187-mm depth in October 2016. Different letters above bars within each figure indicate significant difference (P ≤ .05)

3.2 | Root zone physical properties

The cylinder samples taken in October 2016 showed no effect of type of organic amendment on soil physical properties at 5- to 42-mm depth. At 150- to 187-mm depth, the air-filled and total porosities were higher and the soil density lower in root zones amended with peat than in root zones amended with compost (Table 5).

Establishing greens by sodding instead of seeding resulted in more air-filled and especially water-filled pores and a 26% lower bulk density at 5- to 42-mm depth. In the compost-amended, but not in the peat-amended root zone, an increase in air-filled and total porosity due to sodding was detected even at 150- to 187-mm depth (interaction significant; data not shown).

Turfgrass infiltration rates were not affected by type of organic amendment but were 38 and 54% lower on sodded than on seeded plots 6 and 18 mo after establishment, respectively (Table 5). For hydraulic conductivity, there were significant interactions at both 5- to 42- and 150- to 187-mm depth. Sodding led to increased conductivity of the top layer on peat-amended root zones but had no effect on compost-amended root zones (Figure 2a) The lowest conductivity at 150- to 187-mm depth was found on compost-amended root zones established by direct seeding (Figure 2b).

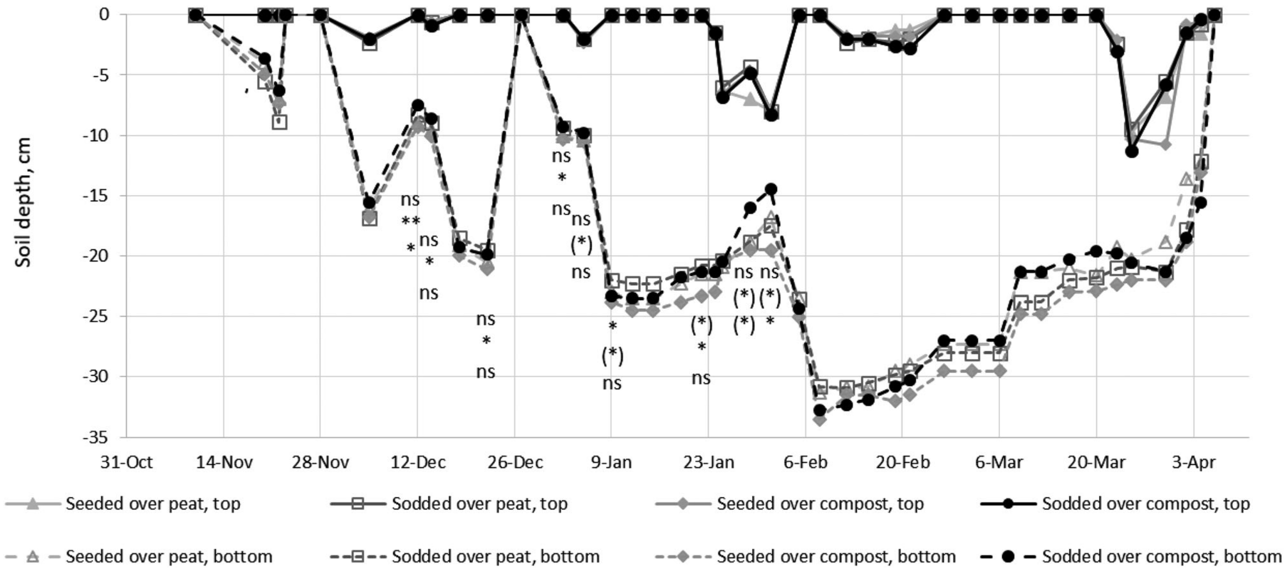

FIGURE 3 Top and bottom of frozen layer in putting green root zones during the second experimental year 2017-18 as affected by seeding vs. sodding and type of organic amendment to the sand-based root zone. $^{**}P \leq .01$, $^{*}P \leq .05$, $(^{*})P \leq .1$; ns, not significant ($P > .10$). Symbols indicate from top to bottom (1) main effect of type of organic amendment, (2) main effect of seeding vs. sodding and (3) the two-factor interaction. No significance symbols indicate no significant difference

Measurements using frost tubes filled with methylene-blue during 2016–2017 showed no difference among treatments in the upper border between frozen and thawed soil (Figure 3). At most measurements in December and January, frost went significantly deeper on seeded and on sodded plots, and on some observation dates, there was also a trend for this difference to be more pronounced on root zones with compost than in root zones with peat (interaction $P < .10$).

3.3 | Collected amount of drainage water and surface runoff

3.3.1 | 2016–2017

On average for treatments, 549 L m^{-2} (91% of the total precipitation of 601 mm) was collected as drainage water during the experimental period in 2016–2017. Until 18 November, drainage rates were significantly (usually around 5%) higher from root zones with peat than from root zones with compost; this continued as a trend ($P < .10$) until 28 December, after which there was no difference between the two root zones (Figure 4a). From January 2017, drainage was higher from seeded than from sodded plot, the cumulative difference reaching a maximum of 47 L m^{-2} on 3 Mar. 2017.

In 2016–2017, only an average of 17 L m^{-2} (3% of the total precipitation) was collected as surface runoff. Most of this was collected when the greens were frozen from early January to mid-February (Figure 4a). When the greens were unfrozen, there was practically no runoff because of the high infiltration capacity. Even the record-high precipitation of 147 mm on unfrozen soil on 5 Nov. 2016 resulted in only 1 L m^{-2} of surface runoff (data not shown). The amount of surface runoff was not influenced by the type of root zone organic amendment or seeding vs. sodding.

Of the total precipitation of 601 mm in 2016–2017, only 35 mm (6%) was not accounted for as drainage or surface water. Most of this was probably lost as turfgrass transpiration during the mostly mild winter without snow cover.

3.3.2 | 2017–2018

During the experimental period in 2017–2018, the average collection of drainage water and surface water amounted to 522 and 310 L m^{-2}, or 55 and 33% of the total precipitation, respectively. The fact that 12% of the total precipitation was not collected as either drainage or surface water was mostly because the top layer of snow above ice-covered greens was removed on 19 and 23 January and 23 March in order to avoid overflow in the collectors for surface water.

As in 2016–2017, drainage rates were higher on peat-amended than on compost-amended root zones until the soil froze up in late November (Figure 4b). At most collections in November and on 20 December there also tended ($P < .10$) to an interaction as seeded turf on peat-amended substrate released more drainage water than the other treatment combinations. During the period with mostly frozen greens from December to April, the cumulative amount of drainage water was always higher from seeded than from sodded greens.

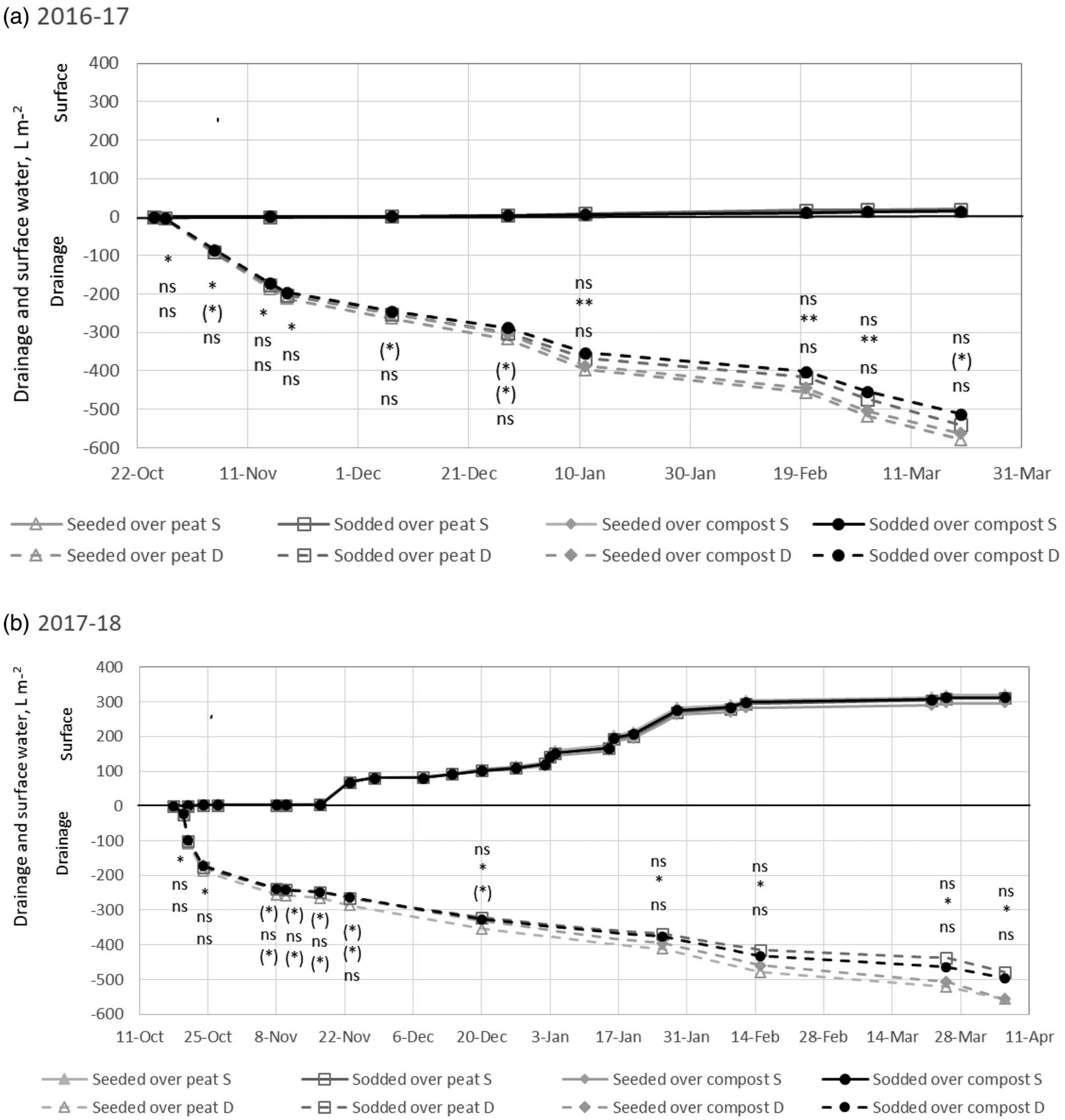

FIGURE 4 Accumulated amount of surface and drainage water during the winters in (a) 2016–2017 and (b) 2017–2018 as affected by experimental treatments. $^{*}P \le .01$, $^{*}P \le .05$, $(^{*})P \le .1$; ns, not significant ($P > .10$). Symbols indicate, from top to bottom (1) main effect of type of organic amendment, (2) main effect of seeding vs. sodding and (3) the two-factor interaction

As in 2016–2017, the amount of surface water was not significantly affected by treatments in 2017–2018. Peaks in surface runoff were recorded between 17 and 23 November (high rainfall on frozen greens) and between 20 and 29 January (mild period with snow melt above ice-covered greens).

3.4 | Fungicide and metabolite detections during the winter 2016–2017

The maximal concentrations of the fungicides and their metabolites in drainage water and surface runoff during the winter 2016–2017 are shown in Table 6. Further

TABLE 6 Main effect of organic amendment, seeded vs. sodded turf, and their interaction on the maximal concentrations of fungicides and metabolites detected in drainage water and surface runoff during the experimental period in 2016–2017

	Boscalid	Pyraclostrobin	Prothioconazole	Prothioconazole-desthio	Trifloxystrobin	Trifloxystrobin acid	Fludioxonil	Fludioxonil metabolite CGA 192155
	μg L^{-1}							
Drainage water								
Peat	0.017	0.005	0.003	0.011	0.002	10.399	0.005	0.571
Compost	0.006	0.005	0.007	0.012	0.003	12.940	0.010	5.343
Significance	ns[‡]	ns	ns	ns	ns	*	ns	***
Seed	0.010	0.005	0.004	0.008	0.003	12.841	0.006	3.000
Sod	0.013	0.005	0.006	0.015	0.003	10.504	0.009	2.914
Significance	ns	ns	ns	***	ns	†	ns	ns
Interaction	ns	ns	ns	ns	ns	†	ns	ns
Date for detection of max. concentration	27 Oct.	27 Oct.	5 Nov.	7 Dec.	27 Oct.	15 Nov.	27 Nov.	11 Jan.
Surface runoff								
Peat	20.2	3.171	0.002	2.079	3.474	6.132	2.653	5.441
Compost	17.8	2.711	0.002	2.776	3.697	7.263	1.326	4.299
Significance	ns	ns	ns	ns	ns	ns	ns	ns
Seed	18.3	3.026	0.002	2.513	3.289	7.434	0.451	0.861
Sod	19.6	2.855	0.002	2.342	3.882	5.961	3.528	8.879
Significance	ns	ns	ns	ns	ns	ns	**	***
Interaction	ns	ns	ns	ns	ns	ns	†	ns
Date for detection of max. concentration	15 Nov.	15 Nov.	3 Mar.	15 Nov.	15 Nov.	15 Nov.	7 Dec.	28 Dec.
ERL[a]	12.5	0.4	0.74	0.033	0.192	320	0.050	100

[†] $.05 < P \leq .1$ ('tendency'); [*] $.01 < P \leq .05$; [**] $.001 < P \leq .01$; [***] $P \leq .001$. [‡] ns, not significant ($P > .1$).
[a] ERL, Norwegian environmental risk limit for aquatic organisms (Norwegian ERL Database, 2019).

information about concentrations at the first sampling after fungicide application and weighed mean concentrations during the entire winter season can be found in Supplemental Table S1.

3.4.1 | Concentrations in drainage water

In 2016–2017, the concentration of fungicides and their metabolites in drainage water were always well below the Norwegian ERL (Table 6). Closest to the ERL was prothioconazole-desthio of which an average concentration of 0.012 μg L^{-1} (ERL = 0.033 μg L^{-1}) was detected on 7 December, about 6 wk after application. The pyraclostrobin metabolite BF 500-6 was barely detected on 15 and 18 November and 7 December, but the average concentration never exceeded 0.0003 μg L^{-1} (data not shown).

Fungicide concentrations were, in most cases, unaffected by the experimental treatments, but for prothioconazole-desthio, the concentrations were higher in drain discharge from sodded than on seeded plots throughout the trial period (Table 6; Supplemental Table S1). As a weighted mean for all samples, they were also higher in drainage water from root zones with compost than from root zones with peat (Supplemental Table S1). The latter effect was even more conspicious for trifloxystrobin acid and especially for the fludioxonil metabolite CGA 192155 of which the maximal concentration on 11 January was almost 10 times higher in drainage water from compost-amended than from peat-amended root zones (Table 6). For this metabolite there was also a significant interaction as the concentation in water samples taken on 18 November, 3 d after application of fludixonil, were three times higher from sod above compost-amended sand than from the the other treatment combinations (Supplemental Table S1).

3.4.2 | Concentrations in surface water

In contrast to the mostly harmless concentations in drainage water, the maximal concentrations of pyraclostrobin, prothioconazole-desthio, trifloxystrobin, and fludioxonil in surface runoff were 10–100 times higher than their respective ERLs (Table 6). The highest concentrations of these compounds, as of boscalid, were found at the first collection of surface water after fungicide application, but for pyraclostrobin, the ERL was exceeded also by the weighed mean concentration for all samples (Supplemental Table S1). The highest concentrations of trifloxystrobin acid and the fludioxonil metabolite CGA 192155, detected on 15 November and 28 December, respectively, did not exceed their respective ERLs (Table 6) The pyraclostrobin metabolite BF 500-6 was barely detected on 3 and 20 March, but the average concentration never exceeded 0.0004 μg L^{-1} (data not shown).

The type of organic amendment to the sand-based root zone had no effect of the concentration in surface runoff for any of the fungicides or their metabolites. Compared with directly seeded plots, sodding led to eight- to tenfold higher maximal concentrations of fludioxonil and its metabolite CGA 192155 on 7 and 28 December, respectively (Table 6). On 28 December, the concentrations of boscalid, pyraclostrobin, trifloxystrobin, and trifloxystrobin acid were also higher in surface runoff from sodded than from seeded plots (Supplmental Table S1).

3.4.3 | Total losses

As a product of water volumes and concentrations, the total losses of fungicides and their metabolite in drainage and surface water during the winter 2016–2017 are displayed in Figure 5. For boscalid, pyraclostrobin, prothioconazole-desthio, trifloxystrobin, and fludixonil, most losses occurred in surface water, 50% or more usually being detected at the first sampling 2 d after application. In contrast, prothioconazole, trifoxoystrobin acid, and the fludixonil metabolite CGA 19155 were mostly lost in drainage water, the period with mostly unfrozen soil from 28 December to 11 January contributing significantly in addtion to the first 3 wk after application. In contrast to the parent fungicides, of which the total losses were always less than 0.2% of the respective application rates, the accumulated losses of the metabolite CGA 192155, amounted to 2.7% of the applied rates of fludioxonil when calculated on a mole basis to account for differences in molecular weight. Correspondingly, the accumulated losses of trifloxystrobin acid were as high as 33.1% of the applied rate of trifloxystrobin (Table 7).

The losses in drainage water were significantly affected by the experimental treatments mainly for prothioconazole-desthio and CGA 192155. Starting in early December, the accumulated losses of prothioconazole-desthio were higher from root zones with compost than from root zones with peat, and higher from sodded than from seeded plots. For CGA 192155, the total losses in drainage water were eight and 13 times higher from compost-amended root zones than from peat-amended root zones on sodded and seeded plot, respectively (interaction significant at $P \leq .05$).

The surface water losses were higher from sodded than from seeded plots for trifloxystrobin acid, fludioxonil, and CGA 192155. Numerically, this was the case also for boscalid, pyraclostrobin, and prothioconazole-desthio, but these differences could not be verfied statistically. Significant interactions for CGA 192155 indicated that the increase in losses in surface water due to sodding vs. seeding was more severe on root zones with peat than on root zones with compost.

3.5 | Fungicide and metabolite detections during the winter 2017–2018

The maximal concentrations of the fungicides and their metabolites in drainage water and surface runoff during the winter 2017–2018 are shown in Table 8. Further information about concentrations at the first sampling after fungicide application and weighed mean concentrations during the entire winter season can be found in Supplemental Table S2.

3.5.1 | Concentrations in drainage water

Although the maximal and mean concentrations of fungicides and metabolites in drainage water were mostly higher in 2017–2018 than in 2016–2017, the ERL was exceeded only for prothiconazole-desthio, most notably at the sampling on 25 March, shortly after soil thaw (Table 8). As in the year before, the maximal concentrations were often higher in drainage water from root zones with compost than from root zones with peat; in 2017–2018, this difference was significant for boscalid, trifloxystrobin acid, and CGA 192155. For prothioconazole and CGA 192155, there were significant interactions indicating that sodding increased the risk for leaching to drains from peat-amended root zones but decreased the risk for leaching to drains from root zones with compost (data not shown).

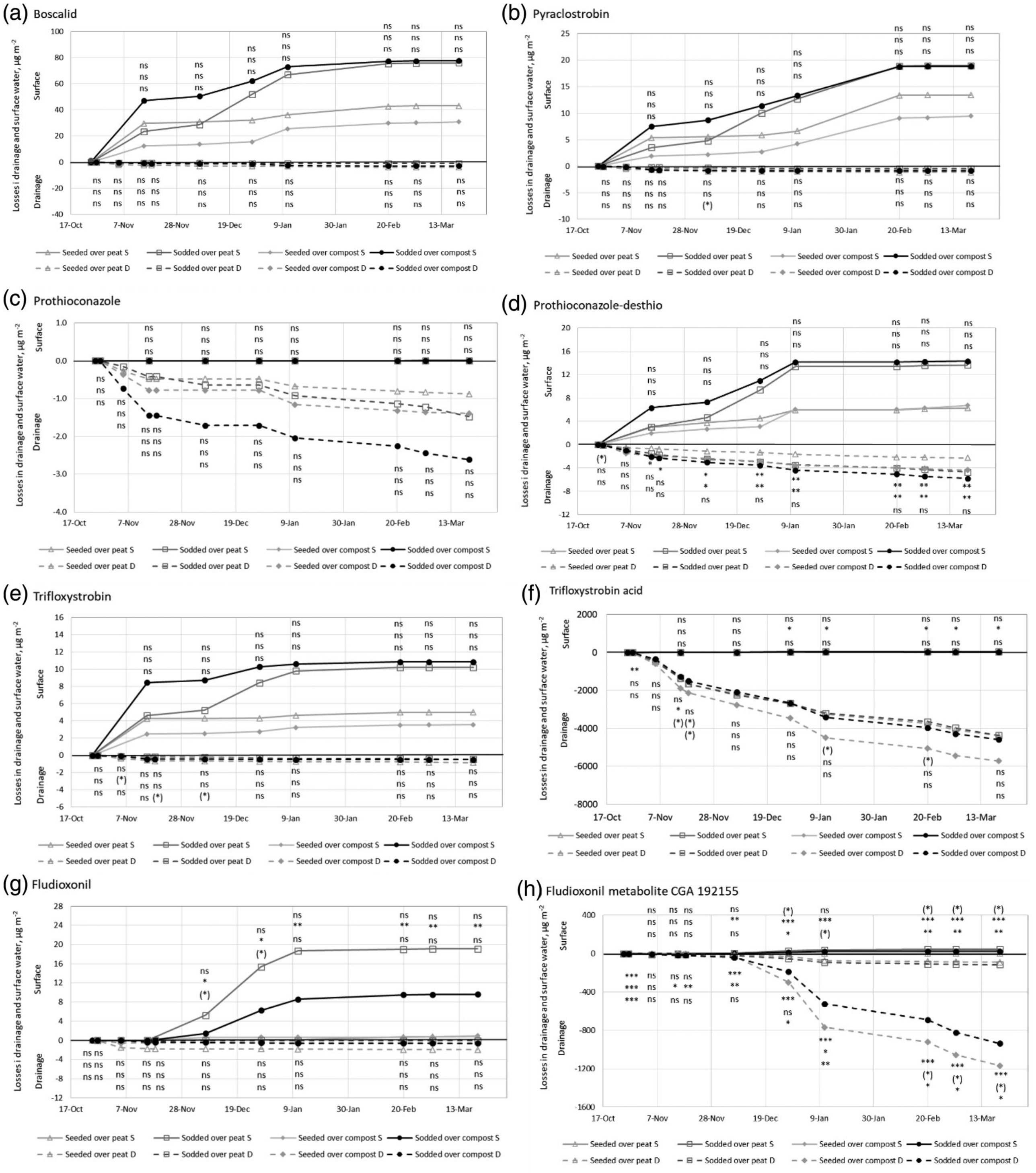

FIGURE 5 Accumulated losses in drainage water (D, negative values) and surface water (S, positive values) of (a) boscalid, (b) pyraclostrobin, (c) prothioconazole, (d) prothioconazole-desthio, (e) trifloxystrobin, (f) trifloxystrobin acid, (g) fludioxonil, and (h) the fludioxonil metabolite CGA 192155 during the winter 2016–2017 as affected by experimental treatments

3.5.2 | Concentrations in surface water

The ERLs for pyraclostrobin, prothioconazole-desthio, trifloxystrobin, and fludioxonil were severely exceeded by the concentrations in surface water during 2017–2018. The most severe incidence was fludioxonil which reached a concentration about 1,000 times higher than the ERL on 17 November (Table 8). However, in spite of the mostly higher maximal concentrations, dilution by the greater surface water volumes in 2017–2018 resulted in lower weighed mean con-

TABLE 7 Total losses of fungicides and the metabolites desthio (of prothiconazole), trifloxystrobin acid (of trifoxystrobin), and CGA 192155 (of fludioxonil) in drainage water and surface runoff and as percent of applied active ingredient in 2016–2017 and 2017–2018. Values are the means of four experimental treatments

		Losses, 2016–2017				Losses, 2017–2018			
	Applied	Drainage water	Surface runoff	Total	Total relative to applied[a]	Drainage water	Surface runoff	Total	Total relative to applied[a]
	μg m^{-2}				%	μg m^{-2}			%
Boscalid	40,000	2.65	56.8	59.45	0.15	26.7	345	372	0.93
Pyraclostrobin	10,000	0.77	15.2	15.97	0.16	0.363	120	120	0.30
Prothioconazole	17,500	1.59	0.005	1.60	0.01	4.95	0.170	5.12	0.03
Desthio		4.26	10.3	14.6	0.09	11.0	22.8	33.8	0.21
Trifloxystrobin	15,000	0.565	7.42	7.99	0.05	0.365	23.3	23.7	0.16
Trifloxystrobin acid		4768	30.5	4,799	33.1	5,846	34.4	5,880	40.6
Fludioxonil	37,500	0.824	7.62	8.44	0.02	1.65	910	912	2.43
CGA 192155		576	22.1	598	2.70	453	84.5	538	2.50

[a]Percent losses were calculated on a mole basis.

centrations of all fungicides and metabolites except fludioxonil in 2017–2018 (Supplemental Table S2) compared with 2016–2017 (Supplemental Table S1).

Except for fludioxonil, which had a 56% higher concentration in surface runoff from peat-amended than from compost-amended root zones on 17 November (Table 8), the experimental treatments had no effect on either maximal or weighed mean concentrations of fungicides and metabolites in surface water in 2017–2018. The pyraclostrobin metabolite BF 500-6 was not detected in either drainage water or surface water at any of the samplings in 2017–2018 (data not shown).

3.5.3 | Total losses

On average for treatments, the losses of fungicides and metabolites were higher in 2017–2018 (Figure 6) than in 2016–2017 (Figure 5). The difference was most prominent for surface runoff, but occurred even for drainage water except for the fludioxonil metabolite CGA 192155. Fungicide losses in surface water were most severe during a period with rain on mostly frozen greens in late November and, particularly for boscalid (Figure 6a), during a period with snow melt on frozen greens in late January. Losses in drainage water were also more severe during the first 3–4 wk after application, but the relatively steady increase in cumulative loss during the winter (Figure 6) also suggests that fungicides and metabolites were lost in drain discharge even in periods with frozen root zones. As in 2016–2017, the losses in drainage water were higher from root zones with compost than from root zones with peat, and higher from seeded than from sodded plots. For the fludioxonil metabolite CGA 192155, there was an interaction as sodding reduced losses on the compost-amended root zone bud had little effect on the root zone with peat.

Unlike in the first experimental year, the losses in surface water were mostly higher from seeded than from sodded plots for pyraclostrobin and prothioconazole-desthio. For fludioxonil, the surface losses were higher on greens established using sod above a peat-amended root zone than for the other treatment combinations.

4 | DISCUSSION

4.1 | Fungicide losses in surface runoff

Our results showed an overall low risk for fungicide and metabolite concentrations in drainage water to exceed the ERLs for aquatic organisms. In contrast, the concentrations of pyraclostrobin, prothioconazole-desthio, and fludioxonil in surface runoff were, on many sampling dates, several orders of magnitude higher than the ERLs. Similar findings have been reported from turfgrass areas in the United States (Bell & Koh 2011; Easton, Petrovic, Lisk, & Larsson-Kovach, 2005; King & Balogh, 2013; Kramer et al., 2009; Petrovic & Easton, 2005; Rice et al., 2010; Slavens & Petrovic, 2012).

The highest fungicide losses were always found in the first runoff collected after fungicide application. In the first year, this did not result in very high losses as the total amount of surface runoff was very low due to the high infiltration capacity of the newly established green. In the second year, the infiltration capacity was still high, yet reduced by, on average for peat and compost-amended root zones, 26% on seeded plots and 46% on sodded plots. While the difference in fungicide runoff between the 2 yr can mostly be explained by what extent the

TABLE 8 Main effect of organic amendment, seeded vs. sodded turf, and their interaction on the maximal concentrations of fungicides and metabolites detected in drainage water and surface runoff during the experimental period in 2017–2018

	Bos-calid	Pyraclostrobin	Prothioconazole	Prothioconazole-desthio	Trifloxystrobin	Trifloxystrobin acid	Fludioxonil	Fludioxonil metabolite CGA 192155
	μg L^{-1}							
Drainage water								
Peat	0.033	0.001	0.015	0.043	0.006	18.316	0.009	0.865
Compost	0.185	0.001	0.016	0.069	0.020	22.662	0.008	3.806
Significance	*	ns[‡]	ns	ns	ns	**	ns	***
Seed	0.150	0.001	0.013	0.039	0.011	21.037	0.013	2.595
Sod	0.069	0.001	0.017	0.073	0.016	19.941	0.003	2.076
Significance	ns	ns	**	ns	ns	ns	ns	†
Interaction	ns	ns	*	ns	ns	ns	ns	*
Date for detection of max. concentration	15 Feb.	26 Jan.	10 Jan.	25 Mar.	17 Nov.	10 Nov.	23 Nov.	26 Jan.
Surface runoff								
Peat	9.390	0.737	0.750	3.063	6.850	3.275	63.325	11.488
Compost	29.600	2.900	0.625	3.213	9.388	2.613	40.538	11.700
Significance	ns	ns	ns	ns	ns	ns	*	ns
Seed	6.940	0.613	0.750	2.588	7.225	2.088	46.875	11.738
Sod	32.050	3.025	0.625	3.688	9.013	3.800	56.988	11.450
Significance	ns	ns	ns	ns	ns	ns	ns	ns
Interaction	ns	ns	ns	ns	ns	ns	ns	ns
Date for detection of max. concentration	20 Oct.	20 Oct.	20 Oct.	20 Oct.	20 Oct.	20 Oct.	17 Nov.	17 Nov.
ERL[a]	12.5	0.4	0.74	0.033	0.192	320	0.050	100

[†] $.05 < P \leq .1$ ("tendency"); *$.01 < P \leq .05$; **$.001 < P \leq .01$; ***$P \leq .001$; [‡]ns, not significant ($P > .1$).
[a]ERL, Norwegian environmental risk limit for aquatic organisms (Norwegian ERL Database, 2019).

surface of the green was frozen, compaction and organic matter accumulation are also likely to have an impact on unfrozen greens which may well have infiltration rates 80–90% lower than in this experiment. Lessons to be learned are therefore, that it is important to maintain a good infiltration capacity to avoid surface runoff, and that fungicide applications shall be avoided if there is a long-term weather forecast for high precipitation rates. American studies conducted during the growing season showed significant reductions in fungicide runoff if the time from application until the first rainfall increased from 12 to 24 h (Branham, Kandil, & Mueller, 2005), and this ‘safety period’ should probably be extended to at least 48 h when applying fungicides at low temperatures in the late fall.

Other aspects to consider when interpreting the high fungicide runoff in this study are that the putting greens only make up a small fraction (usually less the 5%) of the land area of a golf course. Since fungicide applications on Nordic golf courses are usually limited to putting greens, the runoff from the greens will usually be diluted many times before reaching open water. Additionally, it is important to keep in mind that the collectors for surface water in this experiment were placed at the end of each plot without any buffer zone to open water. The Norwegain government recently put into effect a general requirement to 10-m-wide, grass-covered buffer strips to open water for all areas that are sprayed with pesticides and have more than 2% surface inclination (Mattilsynet, 2020). While this regulation is based primarly on agricultural research, there are several reports from the United States showing significant effects of unsprayed buffer zones in turfgrass systems (e.g., Cole et al., 1997; Rice et al., 2017). According to Steinke, Stier, Kussow, and Thompson (2007) it is, nonetheless, questionable to what extent buffer strips reduce runoff in climates where soils are frozen during most of the winter.

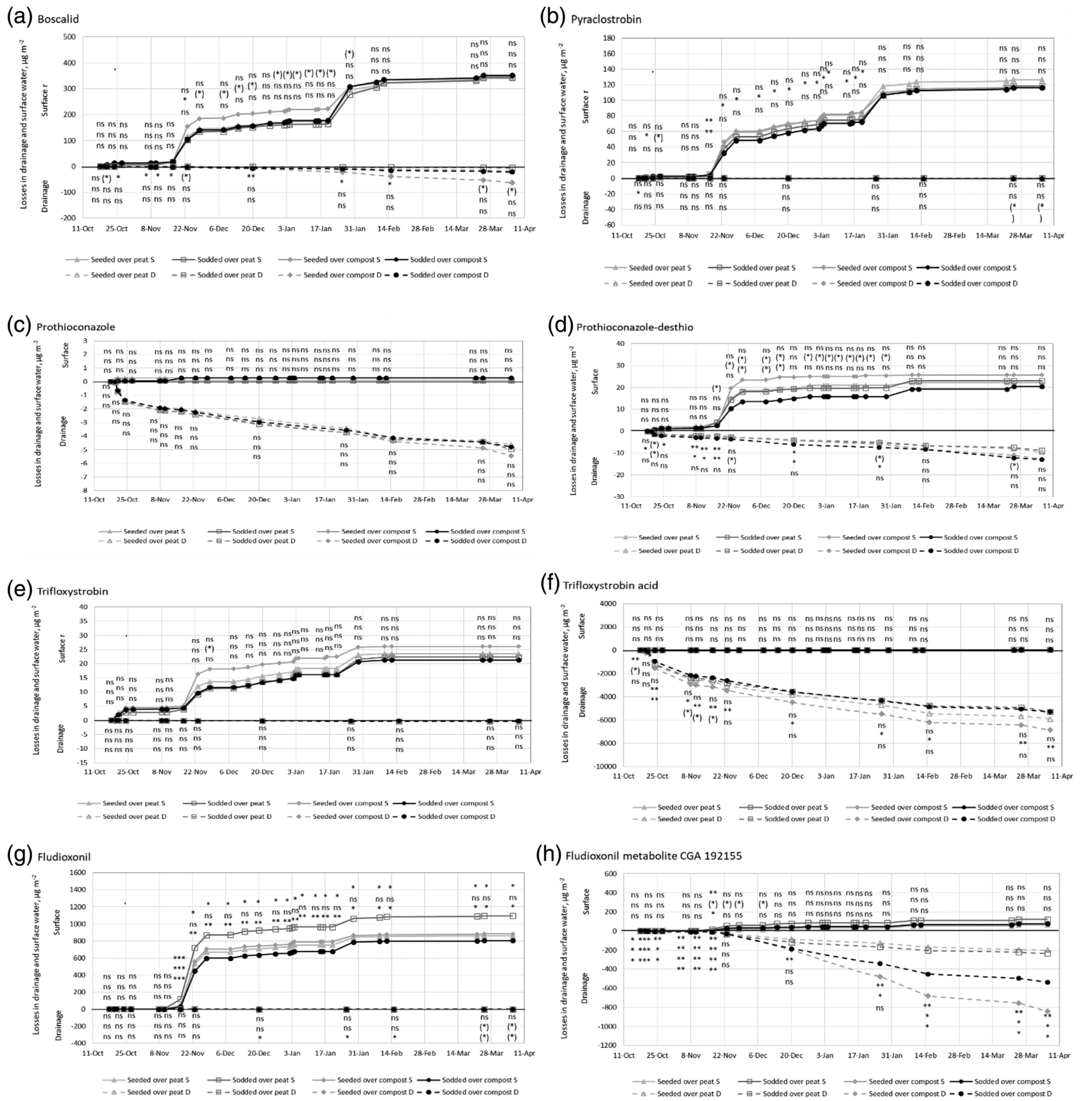

FIGURE 6 Accumulated losses in drainage water (D, negative values) and surface water (S, positive values) of (a) boscalid, (b) pyraclostrobin, (c) prothioconazole, (d) prothioconazole-desthio, (e) trifloxystrobin, (f) trifloxystrobin acid, (g) fludioxonil, and (h) the fludioxonil metabolite CGA 192155 during the winter 2017–2018 as affected by experimental treatments. Significance symbols indicate, from top to bottom: (1) Main effect of type of organic amendment, (2) main effect of seeding vs. sodding and (3) the two-factor interaction

In our experiment, more than 50% of the fungicide runoff occurred on frozen soil in 2016–2017, and more than 90% in 2017–2018.

With the exception of prothioconazole and its metabolite desthio, the maximum concentrations in surface runoff were always higher from sodded than from seeded plots in the first year of our study. The fact that this occurred in spite of higher sorption coefficient is probabaly a reflection that that the mat layer on sodded plots acted as a barrier for contact between the the fungicides and the underlying root zone. A lower infiltration rate combined with higher capillary porosity, and thus a higher water content in the top layer on sodded vs. seeded greens, may also have contributed to this difference as moist and saturated soils are usually most prone to surface runoff (Kenna & Snow, 2000). These result emphasize the importance of a good thatch control program to minimize the risk for fungicide losses in surface runoff. However, in the second year of our study, the impact of the mat layer on fungicide concentrations and total lossses in surface runoff was mostly overrun by the longer period with frozen greens and recurring episodes with snow and ice melt.

4.2 | Fungicide sorption and losses in drainage water

Unlike the other fungicides and metabolites, losses of prothioconazole, trifloxystrobin acid, and the fludioxonil metabolite CGA 192155 mainly occurred in drainage water. Although these compounds were not included in the initial sorption study, one would expect less leaching of fungicides from sodded than form seeded greens, due to the strong sorption to organic matter in the top 3 cm which retained the fungicides from further leaching. This was confirmed for trifloxystrobin acid in the second year of our study and for CGA 192155 in both years. A summary of earlier research found thatch organic carbon to be 40% less efficient than soil organic carbon in retaining pesticides (Carroll, 2008). Carroll and Leshin (2010) later suggested that the thatch decomposition rate, as measured by the N/C or N/C atomic ratio, would be a better predictor for the sorption capacity of a thatch layer than its overall content of organic carbon. While this is probably correct for turf having an undecomposed thatch layer with >7.5% organic C and a bulk density as low as 0.37 kg dm^{-3} as in the modeling approach by Carleton, Sutton, Lin, and Corbin (2009), the sodded plots in our study were rather characterized as having a mat layer, in which case the sorption of fungicides will be proportional to organic C (Dell, Throssell, Bischoff, & Turco, 1994). This is further confirmed by the fact that, after correction for organic C, there was no difference in the sorption coefficient at 0- to 3- and 3- to 30-cm soil depth.

As predicted from the comparable sorption coefficients at 3- to 30-cm depth, we expected to find similar total losses of boscalid, pyraclostrobin, and fludioxonil in drainage water from root zones with peat as from root zones with compost. For prothioconazole-desthio, we expected to find higher losses in drainage water from the compost-amended soil with the higher pH since this metabolite becomes neutral at pH >6.0. These predictions were mostly confirmed by our data which, in addition to prothioconazole-desthio, also showed less leaching of trifloxystrobin acid and CGA 192155 from the peat-amended than from the compost-amended root zone. The leaching of these metabolites would depend on how fast they are produced during decomposition of their parent fungicides. Both fungicides and metabolites are prone to microbial degradation in the soil as well as to aqueous photolysis (Table 2). Pyraclostrobin, prothioconazole, and trifloxystrobin are expected to be rapidly degraded when exposed to sunlight shortly after application to the greens. The metabolite trifloxystrobin acid could also be rapidly degraded by photolysis, whereas prothioconazole-desthio and boscalid are expected to be fairly resistant to photolysis. The lower amounts of trifloxystrobin acid in surface water as opposed to drainage water can be explained by rapid photolysis of trifloxysytrobin acid on the green surface, whereas photolysis was restricted when the metabolite leached below the soil surface. According to the Pesticide Properties DataBase, trifloxystrobin acid (Kfoc 84–194 L kg^{-1}) is much more mobile than the parent trifloxystrobin (Kfoc 1,642–3,745 L kg^{-1}) (PPDB, 2020) and therefore more prone to leaching.

We did not measure the microbial activity, nor the fungicide degradation rates in the root zones, but one could perhaps also expect faster microbial degradation on sodded than on seeded greens due to a denser root system with enhanced microbial activity in the rhizospheres (Aamlid et al., 2009; Torello, 2008). However, this effect may have been counteracted by the stronger sorption capacity of the sod which probably restricted microbial degradation and retained fungicides and metabolites from leaching. The resulting impact would depend on the individual properties of the pesticides and metabolites. As an example, hardly any detection of the pyraclostrobin metabolite BF 500-6, which is produced when pyraclostrobin sorbs to itself to produce a dimer, could probably be ascribed to its strong sorption to the topsoil (Koc = 60,495 L kg^{-1}; PPDB, 2020). The finding that the fludioxonil metabolite CGA 192155 was more prone to leaching from compost-amended than from peat-amended is also compatible with earlier findings that addition of compost enhances microbial activity and thus the fungicide degradation capacity in sand-based putting greens (Aamlid et al., 2009; Strandberg, Blombäck, & Hedlund, 2005). In the second year of our study, enhanced degradation to CGA 192155 also reduced the risk for surface runoff of the far more toxic parent fungicide fludioxonil as demonstrated by the difference between sodded greens on compost-amended vs. peat-amended root zones.

Another important finding in this research was that losses of fungicides and metabolites in drainage water occurred also during periods with frozen root zones. Although the significance of the frost depth measurements in the second year of our study was limited by frost tubes being available only in two out of four replicates, it is an interesting observation that the deepest frost was usually measured in greens seeded on the compost-amended root zone (Figure 3), that is, in the same treatment that had the highest losses of trifloxystrobin acid and CGA 192155 (Figure 6h). This observation is in agreement with earlier reports showing increased pesticide mobility and transport to greater depths in frozen vs. unfrozen soils due to a higher degree of preferential flow through macropores (Holten et al., 2018, 2019; Stenrød et al., 2008). Taken together, these findings emphasize the importance of taking soil freezing and thawing processes into account when modeling the risk not only for surface runoff, but also for leaching of fungicides and metabolites from golf courses in northern areas. Relevant questions are therefore if our results are representative for Nordic golf courses and what measures can be taken to reduce the risk for harmful fungicide concentrations in surface runoff, particularly after application in the late fall to control turfgrass winter diseases.

5 | CONCLUSION

The analyses of surface runoff and drainage water from the putting greens in this experiment showed losses in surface water to be dominated by the parent fungicides. The accumulated losses, on average for 2 yr and expressed as a percent of applied rate, increased in the order prothioconazole < trifloxystrobin < pyraclostrobin < boscalid < fludioxonil. The losses in surface water predominantly occurred when the soil was frozen, as the infiltration capacity of unfrozen soil was very high. In contrast, losses in drainage water were dominated by the fungicides' metabolites due to transformations taking place in the root zone. The highest metabolite losses were found of trifloxystrobin acid followed by the fludioxonil metabolite CGA 192155. Prothioconazole was lost mainly as prothioconazole-desthio in both surface and drainage water. Presumably because of differences in soil pH and frost depths, and despite similar cation exchange capacity, the losses of fungicides and metabolites in drainage water were mostly higher from sand-based root zones amended with compost than from root zones amended with *Sphagnum* peat.

The most important implication of this research is the golf courses that routinely apply fungicides against winter diseases must take necessary precautions to avoid surface runoff, especially of prothioconazole-desthio and fludioxonil due to the high toxicity of these compounds to water-dwelling organisms.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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