

Effect of prescribed fires on the export of dissolved organic matter, precursors of disinfection by-products, and water treatability

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ABSTRACT

In this study, we report for the first time the effect of prescribed fires on the export of dissolved organic matter (DOM) and precursors of disinfectant by-products (DBPs) from periodically (every 2–3 years) and seasonally (i.e., dormant and growing) burned forest fuel materials (i.e., live vegetation, woody debris, and detritus [litter and duff]) and treatability of its rainwater leachate. Periodically applied (every 2–3 years for 40 years) prescribed fires decreased total fuel load ($62 \pm 10\%$), primarily detrital mass ($75 \pm 2\%$). However, functional groups (i.e., phenolic compounds, proteins, carbohydrates, aromatic [1-ring], polycyclic aromatic hydrocarbons [PAHs], and lipids) attached to DOM of ground solid materials did not change significantly. Outside rainwater leaching (from forest fuel materials) experiments showed that the leaching capacity of dissolved organic carbon (DOC) from burned litter samples decreased by $40 \pm 20\%$ regardless of burning season when compared to unburned litter samples. The leaching of total dissolved nitrogen (TDN), dissolved organic nitrogen (DON), ammonium (NH_4^+), and reactive phosphorus (PO_4^{3-}) from burned materials decreased between 40 and 70% when compared to unburned materials. Also, DOM composition was affected by prescribed fire, which partially consumed humic-like substances based on fluorescence analyses. Thus, periodically applied prescribed fires also resulted in a reduction of trihalomethane (THM) ($42 \pm 23\%$) and haloacetic acid (HAA) ($42 \pm 20\%$) formation potentials (FPs), while DOC normalized reactivity of THM and HAA FPs did not change significantly. Additionally, the leaching of N-nitrosodimethylamine (NDMA) precursors, bromide ion (Br^-), and selected elements (K, Ca, Mg, Mn, Fe, S, Na, B, and Al) were not significantly affected by prescribed fires. Finally, coagulant (i.e., alum and ferric) dose requirements and coagulation efficiencies were similar (i.e., removal of DOC, precursors of THMs and HAAs were 52–56%, 69–70%, 78–79%, respectively) in unburned and pre-burned leachate samples.

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1. Introduction

Climate change can result in more frequent and more severe and extended droughts. These conditions may increase the probability of ignition and frequency and severity of wildfires (Bladon et al., 2014; Robinne et al., 2018; Westerling, 2016). The type and amount of surface fuels (i.e., a mixture of live vegetation, woody debris, and detritus [litter (Oi horizon) and duff

(Oe + Oa horizons)]), and their properties, such as moisture and chemical content, impact fuel flammability (Dharssi, 2018). Also, wildfires may greatly impact detrital composition (Coates et al., 2017; Majidzadeh et al., 2015; Wang et al., 2015) which is a primary terrestrial source of dissolved organic matter (DOM) in source waters (Chow et al., 2009; Majidzadeh et al., 2015). Previous studies have shown that post-wildfire runoff increased surface erosion, thus mobilizing sediment, fire-generated materials (i.e., ash and charred), nutrients (i.e., N and P), and trace elements (Abraham et al., 2017; Nyman et al., 2011; Rhoades et al., 2019). Furthermore, wildfires may change DOM composition and concentration of precursors of disinfection by-product (DBPs) (Hohner et al., 2016; Revchuk and Suffet, 2014; Uzun et al.,

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2020; Wang et al., 2016): consequently, treatability requirements for burned watershed-derived DOM can be different when compared to those exported from unburned watersheds (Hohner et al., 2016; Wang et al., 2015; Writer et al., 2014). Considering the widespread use of forested catchments for potable water production (Bladon et al., 2014; Nyman et al., 2011), post-fire rainstorms transporting burned surface fuel leachates to source waters pose a potential concern for drinking water production.

Forest management techniques, such as prescribed fire, are vital tools used to maintain open and fire-resilient forest structures that reduce potentially hazardous wildfire conditions and ignitions (Waldrop and Goodrick, 2012). Although wildfire hazard reduction is one of the primary motivations for prescribed fire use in forest management, it is also employed for many other objectives including but not limited to the control of undesired vegetative species, aesthetics, restoration and maintenance of fire-dependent species and ecosystems, and site preparation (Waldrop and Goodrick, 2012). It is known that prescribed fires may impact forest floor properties (Coates et al., 2020, 2017). Most carbon fluxes come from forest floor leaching in the form of terrestrial DOM (Cronan, 2018), which is one of the main sources of dissolved organic carbon (DOC) and the precursors of DBPs entering source waters (Chen et al., 2019; Chow et al., 2009). Therefore, it is very important to investigate the chemical composition of burned forest materials when considering water quality in source waters. Furthermore, the amount of DOM and its composition are important to consider in conjunction with the potential presence of DBP precursors and their treatability. To date, most studies connecting fire events to changes in source water quality have focused predominantly on wildfires (Chow et al., 2019; Hohner et al., 2017, 2016; Uzun et al., 2020; Wang et al., 2015) due to the severity and widespread impact of wildfires around the globe.

Currently, only a few relevant studies have been conducted to evaluate prescribed fire effects on water quality, especially in the eastern United States (Hahn et al., 2019). Previously, Majidzadeh et al. (2015) performed laboratory burning on the foliar litter of six plant species commonly found in the southeastern United States (US) and evaluated the chemical content and DBP formation potential (FP) of these materials (Majidzadeh et al., 2015). The results showed that the total weight of litter and chemical properties per gram of material (i.e., lignin, nitrogenous compounds, and polysaccharides) were decreased in burned samples because of the loss of organic matter. Also, DOC and chloroform precursor leaching in water decreased in burned materials. To test the effect of prescribed fire on water quality and DBP formation, Tsai et al. (2015) designed another study where litter and duff mixtures from unburned and burned sites (forests consisting mostly of pine trees) were collected and soaked in water. Results were similar (i.e. lower DOC and DBP) for these two studies. Finally, we reported that long-term prescribed fire use may decrease DOC, total dissolved nitrogen (TDN), NH_4^+ , and the FPs of regulated trihalomethanes (THMs [four species]) and haloacetic acids (HAAs [five species]) (Majidzadeh et al., 2019). Although these relevant studies provided useful information, comprehensive studies detailing the effects of dynamic events (i.e., application of burning season, deterioration of the materials under different weathering conditions) on DOM and DBP precursors originating from unburned vs. burned forest materials are still lacking.

Therefore, the objectives of this study were to examine: (i) fire frequency (frequent and repeated prescribed burning [every 2–3 years] for 40 years) and season (dormant or growing season burns) on the export of DOM and selected DBP precursors (regulated: THMs & HAAs, and emerging: chloral hydrate [CH] & N-nitrosodimethylamine [NDMA]) from forest fuels; (ii) the changes in DOM molecular composition; and (iii) the removal of DOM and DBP precursors with commonly used coagulants (i.e., alum

and ferric salts) from leachate waters of forest materials. Prescribed fire reduces potentially hazardous wildland fuel components and enhances habitat for many wildlife species (Waldrop and Goodrick, 2012). Understanding how it may impact the chemistry of burned forest materials, the potential export of DOM and DBP precursors, and their treatability may better inform forest managers and prescribed fire practitioners about the multi-faceted dynamics that should be considered when utilizing prescribed fire to achieve land management objectives.

2. Materials and methods

2.1. Study site and prescribed fires

This study was conducted at the Tom Yawkey Wildlife Center (33.23°N, -79.22°W) in Georgetown, South Carolina (SC), US. The average annual precipitation and temperature in the area were approximately 1400 mm (3.83 mm/day) and 18 °C from 1981–2010, respectively (South Carolina State Climatology Office (SCSCO), 2010). Two paired forest ecosystems primarily composed of longleaf (*Pinus palustris* Miller) and loblolly (*P. taeda* L.) pine, turkey oak (*Quercus laevis* Walter), and sweetgum (*Liquidambar styraciflua* L.) (Coates et al., 2017, 2018a) were located at this study site and differed in their long-term management strategies.

Unmanaged Site A had not been burned or harvested from 1978 to 2014. Site B had been burned every 2–3 years over that same time period. The most recent burn had been performed in 2014 (Site B) (Fig. 1). To accomplish our research objectives, composite fuel samples (forest materials: live vegetation; woody debris; detritus [litter and duff]; ash and charred materials [burned sites only]) were collected from the unmanaged site (Site A) and the previously burned sites (Site B) before additional seasonal prescribed burns were conducted in 2015. In 2015, six units (0.01–0.02 km² each) in Site B were burned: three units during the dormant season (Mar. 9–11, 2015) and three units during the growing season (May 5–6, 2015). The prescribed burns represented typical dormant and growing season ignitions implemented by the SC Department of Natural Resources at the Tom Yawkey Wildlife Center to reduce potentially hazardous wildland fuels, maintain open forest conditions, and improve wildlife habitat for the red-cockaded woodpecker (*Dryobates borealis* Vieillot). All burns were head-fires and mean flame lengths in each of the burns were approximately 0.3–1.0 m, which is characteristic of low intensity, low severity fires in this region (Coates et al., 2018a). Fire temperatures were recorded using in situ thermocouple-datalogger assemblies. Average peak burning temperatures ranged between 200 and 315 °C (Coates et al., 2018b).

2.2. Material collection and leaching experiment design

Sample collection frames (1 m x 1 m) were placed on the ground every 50 m along a 300 m linear transect within each treatment unit (Fig. 1). All live and dead vegetation, fine woody debris (≤ 7.62 cm diameter) (Woodall and Liknes, 2008), litter, duff, ash, and charred materials (when present) were collected within these frames. Following Site B prescribed burns, this represented a mixture of both burned, partially charred, and unburned materials due to the nature of the low intensity and severity prescribed burns. Collected materials were oven-dried at 70 °C for no less than 48 hrs immediately after collection to obtain oven-dry fuel mass. Composite fuel samples were randomly selected from each of the following treatment (prescribed burning applied) units for additional experiments: Site A unmanaged, Site B pre-burned, Site B dormant season post-burn, Site B growing season post-burn. One kg mixtures of collected materials from the unmanaged and burned plots were packed in aluminum screens (1 mm x 1 mm

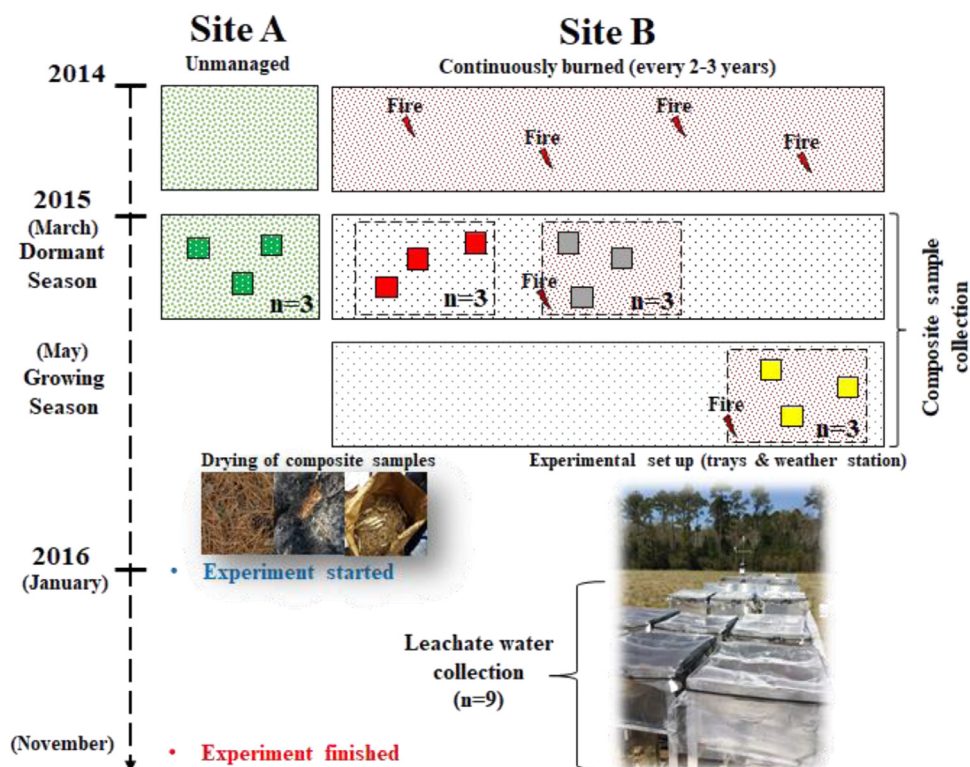


Fig. 1. Scheme for the forest management strategy and experimental design. Site-A and Site-B represent unmanaged (unburned) and managed (burned) areas, respectively. Square sampling frames (1 m x 1 m) where the composite fuel samples were collected were selected randomly for each site. Color assignments: green, red, dark gray, and yellow for unmanaged, managed pre-burned, after dormant season and after growing season samples, respectively. n: shows the number of samples collected.

mesh size). These wrapped samples were then individually placed in custom-made open top aluminum trays (0.6 m x 0.6 m x 0.3 m). An empty control tray was also placed in the open field with the sample trays to capture unimpeded rainwater (Figs. 1 and S1). A weather station was located next to the trays, which recorded precipitation, air temperature, pressure, humidity, and wind speed every 15 min (Fig. 1).

2.3. Analytical methods

2.3.1. Pyrolysis analysis with ground solid samples

To examine the effect of prescribed fires on the chemical structure of detritus materials, ground solid materials were subjected to pyrolysis gas chromatography-mass spectrometry (Py-GC/MS). Samples were oven-dried for no less than 48 hrs and ground using a Wiley mill (2 mm). One to two mg of these materials were then inserted into quartz tubes for chemical analysis, following methods described elsewhere (Coates et al., 2017; Song and Peng, 2010).

2.3.2. Leachate water collection

For nearly a year, when precipitation fell over the trays in the field, leachate water of each tray was collected in glass carboys (total volume of each carboy: 23 L) located underneath each tray as rain saturated the material and drained through the trays (Figure S1). The amount of daily precipitation and the volume of tray leachates were recorded for each rain event (Figures S2 and S3). The ability to hold water was not significantly different ($p > 0.05$) for each treatment, where the difference was $< 7\%$ in total volume collected in the trays (Figure S3). After leachate water collection, samples were transferred into pre-cleaned 1L amber bottles, immediately filtered with pre-burned and pre-washed glass filters (Whatman 934-AH, $\sim 0.7 \mu\text{m}$ pore sized), and kept in the

refrigerator at $4 \text{ }^\circ\text{C}$ until water quality and DBP tests were conducted within one week after collection. During the leachate water collection period, temperatures dropped below freezing only a few times, and the average temperature ($19 \text{ }^\circ\text{C}$) between Jan. 1 and Nov. 1, 2016, approximated the historical, (1981–2010) average temperature of $18 \text{ }^\circ\text{C}$ for this region (Figure S4). Several lengthy, wet periods were observed, but only two relatively dry periods occurred when the average precipitation was less than the historical, average precipitation of 3.83 mm/day : Mar. 15–May. 30, 2016 (1.84 mm/day) and Oct. 1–Nov. 30, 2016 (0.02 mm/day except for three rainy days).

A total of 33 rain events producing a considerable amount of leachate water were recorded, but detailed measurements were conducted for nine selected events: three initial flushes (Jan. 15, Jan. 23, and Feb. 5); three events during relatively dry periods (Mar. 15, Mar. 28, and May 14); the first significant flush after a dry period (May 31); one event during a relatively wet period (Aug. 12); and an additional event following a dry period (Nov. 14). The export of DOM never reached zero and the total volume was collected in the trays, therefore, concentrations of selected parameters (i.e., DOC, TDN, dissolved organic nitrogen [DON], and dissolved reactive phosphorus [PO_4^{3-}]) for 24 unexamined rain events were approximated using linear interpolation. Cumulative exports were calculated using the following formula: $C_1 \times V_1 + C_2 \times V_2 + \dots + C_n \times V_n$, where C and V were measured (for nine samples) or calculated (for 24 samples) concentrations (mg/L) and volumes (L) of the samples collected during each rain event, respectively. The trays were open-top, therefore, bird feces or other potential contaminants were removed in the few instances they were detected. Due to the nature of this field experiment, it was not possible to completely exclude the impacts of these unwanted materials, especially in nutrient concentrations.

2.3.3. Water quality, DBP and water treatability tests

During the sample collection period, when enough leachate water (≥ 2 L) was collected in carboys, water quality parameters (i.e., DOC, TDN, DON, ammonium, nitrate [NO_3^- -N], nitrite [NO_2^- -N], PO_4^{3-} and bromide ion [Br^-]); spectroscopic properties (i.e., UV_{254} , E2/E3 [inversely correlated with a molecular weight of DOM (Peuravuori and Pihlaja, 1997)]; humification index [HIX]; fluorescence index [FI] and freshness index [$\beta:\alpha$]); fluorescence excitation-emission matrices (EEMs) (I: tyrosine-like, II: tryptophan-like, III: fulvic acid-like, IV: soluble microbial byproduct-like and V: humic acid-like) (Zhou et al., 2013); concentrations of selected dissolved elements (K, Ca, Mg, Mn, Fe, S, Na, B, and Al); and DBP (THMs, HAA₅, CH, and NDMA) FPs were measured. SUVA_{254} (L/mg-m) (describing the DOM aromaticity) was calculated with the following formula: $(\text{UV}_{254}/\text{DOC}) \times 100$. When the collected leachate water was insufficient (< 2 L) for all water quality and DBP tests, it was discarded, and carboys were cleaned immediately.

DBP FP tests were conducted in the presence of excess oxidants for an extended contact time (five days) to measure the maximum formations in the samples. Initially, pH was adjusted to 7.8 with 20 mM phosphate buffer to mimic typical finished water pH levels in drinking water treatment plants (DWTPs). For THM, HAA, and CH FP tests, initial oxidant (chlorine [Cl_2] stock [$\sim 5\text{--}6\%$ available Cl_2]) spiking doses were calculated by using the formula: $3 \times \text{DOC} + 7.6 \times \text{NH}_4^+ + 10$ (Krasner et al., 2004). After five days of reaction at room temperature (21–22 °C) in 125 mL amber bottles (headspace free), residual Cl_2 (always > 10 mg/L) was measured and quenched with a slightly higher dose of ascorbic acid than stoichiometrically required. NDMA FP tests were performed with spiking pre-formed monochloramine (NH_2Cl) stock solution ($\text{Cl}_2:\text{N} = 4:1$ by weight) prepared by the slow (drop by drop at pH 9.0) addition of Cl_2 into the ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) solution (Jones et al., 2012). Initial NH_2Cl doses applied to the samples were 100 mg/L. After 5 days of contact time at room temperature, residual oxidants (always > 40 mg/L) were measured and quenched with a slight excess amount of sodium thiosulfate than stoichiometrically required. Finally, samples were extracted and analyzed with GC-ECD and GC MS/MS for THM, HAA and CH, and NDMA, respectively. Concentrations of free and total chlorine were determined following Standard Method 4500-Cl F (APHA/AWWA/WEF, 2012).

Selected unfiltered leachate water samples were treated with alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) and ferric chloride (FeCl_3). Before each water treatability test, preliminary jar test experiments were performed at $\text{pH} 6.0 \pm 0.1$ (a varying amount of carbonate buffer, NaOH, and HCl were used as needed) with six doses (10, 15, 20, 30, 40, and 50 mg/L for alum, and 5, 7.5, 10, 15, 20, and 25 mg/L for FeCl_3) of coagulant (a typical enhanced coagulation practice) to determine the optimal DOC, UV_{254} , and turbidity removals. Based on the results, optimum removal conditions (highest removals reached with the lowest coagulant dose) were selected (Figure S5). Then, additional tests (i.e., DOC and DBP FP removals) were conducted with the treated waters under pre-selected optimum treatment conditions and percent removal efficiencies were calculated with the following formula: $(C_0 - C_{\text{tre}})/C_0 \times 100$, where C_0 and C_{tre} are concentrations of raw and treated waters, respectively. Detailed descriptions for general water quality, optical properties & fluorescence spectroscopy, analysis of DBPs (sample extraction and quantification), treatability tests, analytical methods and minimum reporting levels (MRLs) (Table S1) were presented elsewhere (Majidzadeh et al., 2019; Ruecker et al., 2017; Uzun et al., 2020, 2018), and in the Supporting Information (SI). A two-tailed student's *t*-test with a significance level of 0.05 was performed to document statistically significant differences where necessary.

3. Results and discussion

3.1. Prescribed fire alters fuel mass distribution

Fuel height in both the unmanaged and pre-burned sites was 30–50 cm (Coates et al., 2017). However, total fuel mass (kg/m^2) of composite fuel samples was significantly higher in the long-term unmanaged Site A ($4.48 \pm 0.11 \text{ kg}/\text{m}^2$) ($p < 0.05$) than the frequently burned Site B (pre-burned) ($1.73 \pm 0.51 \text{ kg}/\text{m}^2$) (Figure S6). Live fuels were not detected in the samples collected after seasonal prescribed fires because they were consumed during the prescribed fires. Fine woody debris mass (≤ 7.62 cm diameter) was similar ($p > 0.05$) for both unmanaged and managed sites, regardless of the burning season. Fine woody debris is often quite heterogeneous across a given landscape, often at a resolution as small as 0.25 m^2 in frequently burned pine systems (Loudermilk et al., 2017). Often, fast-moving, low intensity, low severity surface fires may not completely consume woody debris ≤ 7.62 cm in diameter. These fuel particles may only be partially charred, therefore, significant differences between burned and unburned forests or pre- and post-fire masses within burned forests may not be detected. This was observed because of the burns at the Tom Yawkey Center where incomplete combustion of woody debris resulted from the prescribed fires. Prescribed fires may also increase the smallest size class of woody debris (< 0.64 cm diameter) immediately post-fire due to mortality of live, understory vegetation during a burn event. This may have been reflected in the woody debris results for the pre-fire masses in Site B, as well. It should be noted that larger woody debris, > 7.62 cm diameter were not included, therefore inferences about those larger fuels cannot be made as a result of this study.

Detritus mass ($3.68 \pm 0.47 \text{ kg}/\text{m}^2$) in unmanaged Site A was significantly greater ($p < 0.05$) than Site B ($0.89 \pm 0.10 \text{ kg}/\text{m}^2$). This indicated the continuous consumption of detritus as a result of frequent prescribed fires in Site B. Since pre-burned materials were collected from Site B about 1 year after (2015) the last prescribed fire (2014), higher detritus mass ($0.89 \pm 0.10 \text{ kg}/\text{m}^2$) was observed compared to mass that was collected right after seasonal burns (0.20 ± 0.05 , $0.52 \pm 0.11 \text{ kg}/\text{m}^2$ for dormant and growing season burns, respectively). This can be associated with the accumulation of litter between 2014 and 2015 and the consumption of such materials during seasonal prescribed burns. Consequently, the combination of fuel mass changed (more woody substances and less detrital materials) per unit weight (kg) of the materials collected from burned sites. These observations further support that prescribed burning consumes detritus materials, changing both their mass and depth (Coates et al., 2017; Cronan et al., 2015).

3.1.1. Prescribed fire does not alter functional groups attached DOM

Phenolic compounds (i.e., lignin-like) constituted about 76% (10% higher than unmanaged samples) of all burned samples, regardless of management, which can be related to incomplete combustion of the materials during prescribed fires (Coates et al., 2017). In contrast to the significant decrease in detrital mass, prescribed burning did not significantly change the detrital chemical functional groups (proteins [nitrogen compounds], carbohydrates [oxygen compounds], and lipids [aliphatic compounds]) (Figure S7). The aromatic compounds were found in higher concentrations in unmanaged samples; a higher level of toluene was observed in unmanaged samples compared to burned samples (Coates et al., 2017). Previous studies reported conflicting results on the formation and fate of polycyclic aromatic hydrocarbons (PAHs) after burning. While some researchers reported an elevated level of PAH formation which survived and remained for a long period (Forbes et al., 2006; Vergnoux et al., 2011a), others indicated that PAHs are transported into the soil quickly. In only a few months

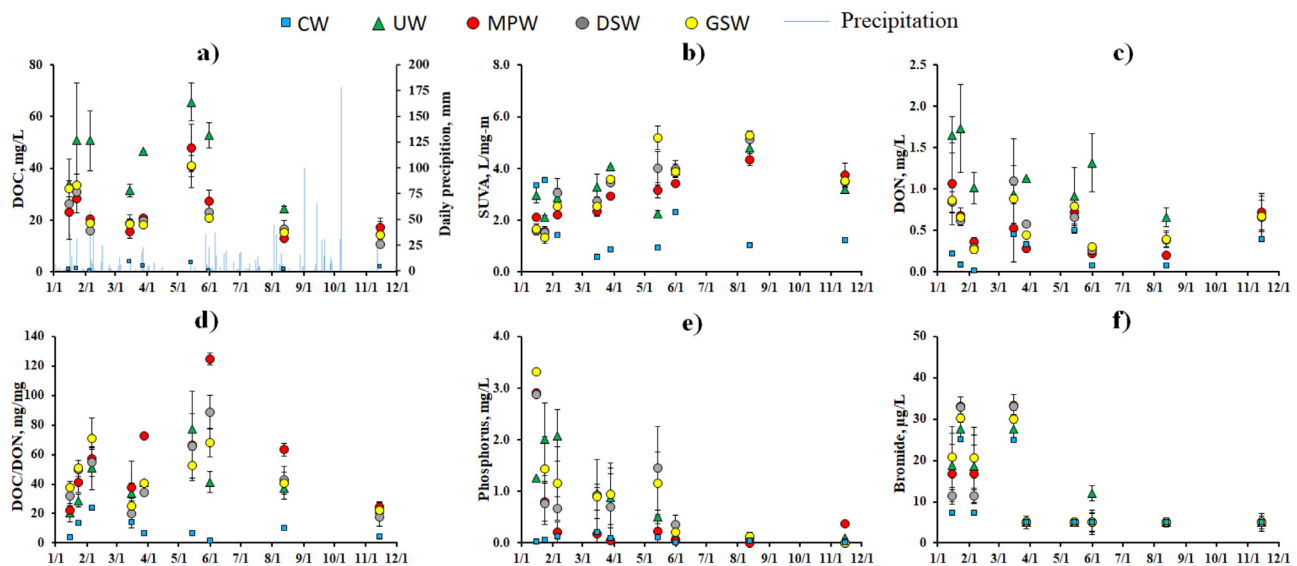


Fig. 2. DOC (a), SUVA₂₅₄ (b), DON (c), DOC/DON (d), P (e), and Bromide (Br⁻) (f) concentrations for unmanaged and managed sites' leachate waters. CW: Control water (rainwater), UW: Unmanaged leachate water, MPW: Managed pre-burned leachate water, DSW: After dormant season leachate water, GSW: After growing seasonal leachate water. Error bars show standard deviation between three replicates. If measured Br⁻ concentration < minimum reporting level (MRL) (10 µg/L), values were set 5 µg/L to complete profiling.

post-fire, PAH levels may return to pre-fire levels (Kim et al., 2003). In our study, PAHs constituted only 2–3% of the total biomass, regardless of the treatment (Figure S7). This indicated that the effect of prescribed burning on PAH formation was minimal, possibly due to incomplete combustion of the detritus resulting from low-intensity burning (Chen et al., 2018; Coates et al., 2017).

3.2. Prescribed fire affects water quality: carbon, nitrogen, and phosphorus release

The average DOC value was minimal (1.64 ± 1.15 mg/L) for rainwater collected in control trays (control water, CW). For litter trays, DOC values were 41.3 ± 15.8 mg/L, 23.7 ± 10.4 mg/L, 22.5 ± 9.1 mg/L, and 23.5 ± 9.4 mg/L in leachates from unmanaged water (UW), managed pre-burned water (MPW), post-dormant season burn water (DSW), and post-growing season burn water (GSW), respectively. Similarly, as shown in Fig. 2a, during the initial seven samples (initial flushes [$n = 3$], dry period [$n = 3$], and the first flush after dry period [$n = 1$]), average DOC releases from unmanaged samples (45, 48, 53 mg/L) were significantly higher ($p < 0.05$) than DOC releases from other burned leachate samples (24–28, 26–28, and 21–27 mg/L for MPW, DSW, and GSW, respectively). After this period, significantly ($p < 0.05$) lower DOC concentrations were measured in all samples (UW 24 mg/L, and others 13–17 mg/L) during the relatively wet period sampling (Aug. 12), and finally, the levels were similar (11–17 mg/L) for the last samples (Nov. 14) regardless of management (Fig. 2a). DOC leaching trends during initial flushes in relatively colder periods indicated gradually decreased leaching capacity of material for all treatments. However, opposite trends for the relative drier and warmer periods indicated that microbial activity may have played a role in enhanced DOC export from all litters. Overall, cumulative DOC export values (16.9 gram for 400 L) from unmanaged materials (8.9 gram for 380 L) were significantly higher ($p < 0.05$) than the export values from managed samples (Fig. 3a). Results indicated that long-term prescribed burning can decrease DOC leaching capacity of forest litter (Majidzadeh et al., 2019, 2015), and burning season does not significantly ($p > 0.05$) affect this result. This might be related to loss of carbon during the burning process (Hongve, 1999; Vergnoux et al., 2011b) and consumption of

materials (i.e., detritus) that have a relatively higher potential of DOC leaching (Zhang, 2017), particularly detrital mass. However, it should not be forgotten that these results were obtained from burned and unburned substances and their leachates. Other natural factors (hydrology, geology, soil properties, erosion, etc.) not accounted for by the tray design play a significant role in DOM export.

During the experiment, average SUVA₂₅₄ values showed fluctuations in the control water. However, those were comparable for all leachate water samples (2.9–3.3 L/mg-m), regardless of the treatment (Fig. 2b). This indicated that these prescribed fires did not likely change overall aromaticity (Krasner et al., 1996) of the leachate water derived from the litter materials. However, in general, higher SUVA₂₅₄ values were observed as time progressed. This indicated that the leaching rate of non-aromatic compounds may have been faster than the leaching rate of aromatic compounds, regardless of management strategy. Opposing trends were observed for samples collected after a long dry period (between Mar. 28 and May 14) from the unmanaged leachate. As indicated before, this period was dry, and the air temperature was relatively warm (18 °C). Previous studies indicated that decomposition of newly generated compounds is related to charring intensity and fuel source (Baldock and Smernik, 2002; Coates et al., 2017; Czimczik et al., 2003; Masiello et al., 2002). Thus, our observations indicated that newly formed phenolic compounds and PAHs in the burned samples were not significantly affected by long dry periods and continued to release from burned litters as time progressed.

In general, NO₂⁻ and NO₃⁻ concentrations were governed by CW and values were mostly lower than minimum reporting levels (MRLs) (data not shown). However, TDN, DON, and NH₄⁺-N concentrations were mostly higher in unmanaged leachate water than burned leachate waters, ranging from 1.03 to 2.72, 0.72 to 1.73, and 0.13 to 1.80 mg/L, respectively (Figs. 2c, S8a, and S8b). Like DOC trends, the cumulative export of DON and TDN were significantly higher in unmanaged samples (Fig. 3b and c). It has been shown that at temperatures of 200–500 °C, nitrogen volatilizes, and most of the organic matter is released to the atmosphere (Knicker, 2007; Waldrop and Goodrick, 2012). Thus, decreases in nitrogen concentrations in burned litter samples can primarily be related to the

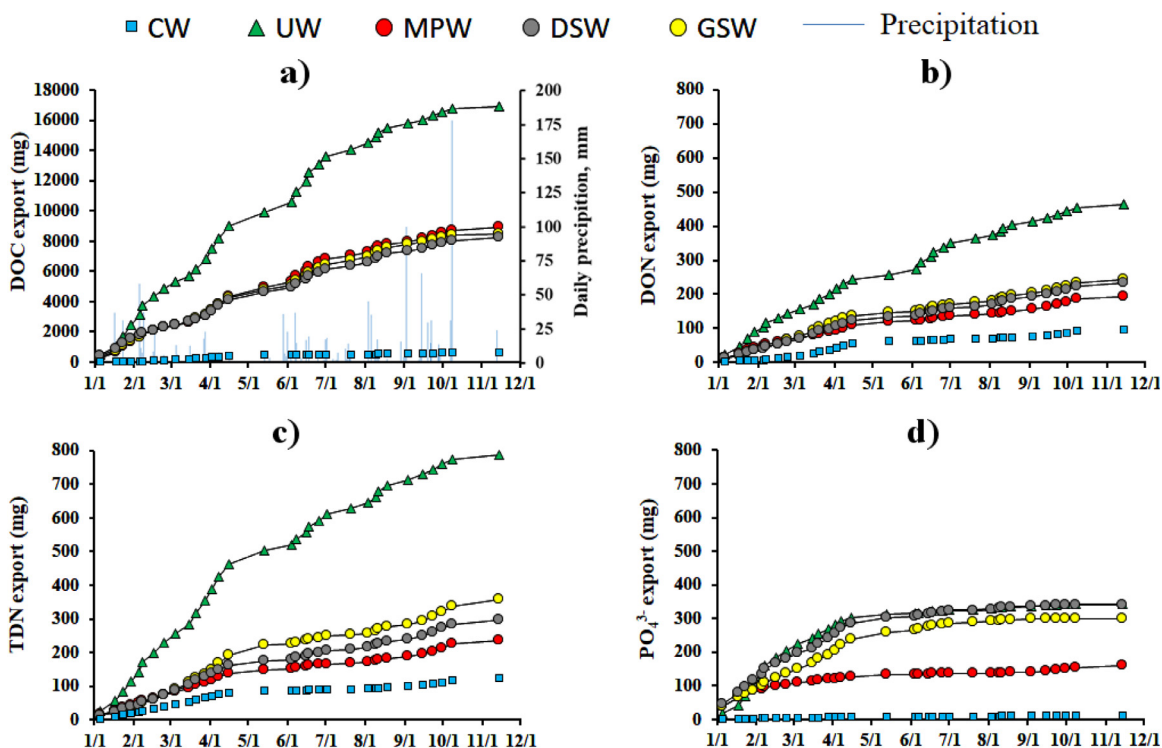


Fig. 3. Cumulative export for DOC (a), DON (b), TDN (c), and reactive phosphorus (PO_4^{3-}) (d) for unmanaged and managed sites' leachate waters. CW: Control water (rainwater), UW: Unmanaged leachate water, MPW: Managed pre-burned leachate water, DSW: After dormant season leachate water, GSW: After growing seasonal leachate water.

volatilization of nitrogen during combustion (Majidzadeh et al., 2015). Finally, the ratios of DOC/DON in all samples were mostly comparable except for the first significant flush after a dry period (May 31) (Fig. 2d), when DOC/DON was considerably higher in burned samples. This may indicate that DON leaching in burned materials is more sensitive to dry conditions.

Phosphorus leaching from burned materials was higher than leaching from unburned samples during the initial six samples (Fig. 2e). The values reached similar levels (<0.4 mg/L) after the initial sampling was completed. Therefore, the cumulative export of phosphorus in UW, DSW, and GSW samples was comparable (Fig. 3d), however, lower concentrations were measured in managed MPW. It has been shown that burning can cause the short-term release of phosphorus, which can be consumed quickly by live vegetation (Zhang, 2017). Relatively higher concentrations for post-dormant and growing season burns can be associated with fresh burning and the release of PO_4^{3-} during fires. However, decreased PO_4^{3-} concentrations in MPW samples can be associated with consumption of freshly released PO_4^{3-} by live vegetation between the latest fire application date (2014) and the sample collection date (2015). However, these results showed that continuous management with prescribed fire may decrease PO_4^{3-} concentrations significantly (up to 60%) by the subsequent fire-initiated release and live vegetation consumption processes.

3.3. Prescribed fire does not affect the release of bromide and other elements

The concentration of Br^- in all leachate water samples varied over time and was mostly governed by CW. For the initial four samples (Jan. 15 and 23, Feb. 5, and Mar. 15), bromide concentration in CW was 16 $\mu\text{g/L}$, and in all other samples, it was 22–25 $\mu\text{g/L}$ (Fig. 2f). Following these initial samples, bromide concentrations decreased sharply and most were under MRL (10 μL , Ta-

ble S1). Previously we indicated that the release of bromide from burned materials could be elevated due to high-temperature combustion (Wang et al., 2015). However, in this study, bromide concentrations in unmanaged and managed litter leachates were similar. This showed that low intensity, low severity prescribed fires are not likely to affect bromide release from litter samples.

It has been shown that fires can cause more trace element fluxes than other types of land-use changes, but the effect is element-specific (Abraham et al., 2017b; Potthast et al., 2017). After prescribed fires, some parameters (electrical conductivity [EC], P and K+) recovered fast (1 year), but some others (pH, C, N, Ca^{2+} and Mg^{2+}) took more time for recovery in the natural environment (Alcañiz et al., 2016). Particularly high-intensity fires can cause the volatilization of elements (DeBano, 1990). Since high temperatures are required for this process, no (i.e., Mg^{2+} , Ca^{2+} , and Mn [DeBano, 1990]) or limited (i.e., K and S [DeBano and Conrad, 1978]) effects of high-intensity fires on element volatilization have been previously reported. In this study, during the sample collection period, dissolved concentrations of elements did not show a significant difference between samples, except Mn for unmanaged leachate waters (Figure S9). In general, concentrations increased during the long dry period (Mar. 15–May 30), indicating the accumulation of these elements in the litter. Then, during the wet rainy period (May 1–Nov. 15), most of the accumulated elements were flushed out of the samples. Taken together, these results showed that prescribed fires at the Tom Yawkey Center did not affect the leaching capacity or processing of trace elements from the forest materials significantly.

3.4. Prescribed fire consumes terrestrial-derived DOM

It has been shown that EEM regions and fluorescence indices can provide useful information about DOM (Fellman et al., 2010), and fires can cause changes in these parameters (Hohner et al., 2016; Revchuk and Suffet, 2014; Uzun et al., 2020). Despite the

Table 1
Comparison of EEM regions and fluorescence indices of leachate waters.

Samples (n = 6)	EEM Regions				Fluorescence Indices			
	I + II (Protein-like)	III DOC-eq (mg/L)	IV (Fulvic-like)	V DOC-eq (mg/L)	FI (SMP-like)	E2/E3 DOC-eq (mg/L)	HIX (Humic-like)	β/α DOC-eq (mg/L)
UW	7.58 ± 3.89	6.55 ± 2.79	10.67 ± 6.11	11.91 ± 6.22	1.40 ± 0.05	5.99 ± 0.19	6.68 ± 2.85	0.50 ± 0.02
MPW	7.72 ± 4.12	7.06 ± 3.62	7.66 ± 4.16	8.88 ± 3.65	1.41 ± 0.11	5.58 ± 0.09	4.10 ± 1.45	0.52 ± 0.05
DSW	5.67 ± 3.01	4.84 ± 2.36	7.00 ± 3.72	7.74 ± 3.77	1.41 ± 0.12	5.58 ± 0.11	5.22 ± 1.61	0.56 ± 0.07
GSW	5.50 ± 2.50	4.91 ± 1.85	6.67 ± 3.80	7.35 ± 3.59	1.41 ± 0.12	5.57 ± 0.15	5.22 ± 1.61	0.56 ± 0.07

UW: Unmanaged leachate water, MPW: Managed pre-burnt leachate water, DSW: After dormant season leachate water, GSW: After growing seasonal leachate water. Protein-like indicates tyrosine-like + tryptophan-like. Errors indicate standard deviation of six samples (n indicates the number of data used for calculations) measurement for each sample. DOC-eq. (equivalence) calculated by: (percent [%] volume of the region/100) x DOC (mg/L) of the sample.

interference of iron reported previously (Hohner et al., 2016; Homann et al., 2011), the effect of iron interference is expected to be minimal in our samples due to low levels (<0.11 mg/L) (including the effect of CW). During the experiment, we selected six dates (Jan. 15 and 23, May 14 and 31, Aug. 12, and Nov. 14) representing all previously stated distinct periods and EEM regions and fluorescence indices of DOM in leachate waters were measured. The data (Table 1) indicated that repeated burning (every 2–3 years) could decrease fluorescence intensities, particularly SMP- and humic-like DOC-eq. (calculated as: percent [%], volume of the region/100 x DOC [mg/L]) in DOM. This supported lower DOC concentrations in pre-burn samples compared to unburned samples. Furthermore, the data showed that fresh burning (regardless of season) can cause further decreases in all EEM DOC-eq. regions and consume particularly humic-like substances. Right after a dry period, however, all EEM regions of DOM increased in UW and MPW samples, which might be related to the combination of both biotic and abiotic factors in trays (Figure S10). However, we cannot provide detailed information as these factors spanned beyond our research purposes. A substantial reduction in all regions was observed during the wet period. This pattern was obvious in all samples, but particularly noticeable in UW and MPW samples, indicating the decreased leaching potential of all materials. Finally, HIX indices also provided information about the DOM source, where HIX > 6 indicates terrestrial DOM, and HIX < 4 implies biological or aquatic origin (Kalbitz, 2001). Our results showed that prescribed fires can consume particularly terrestrial-derived DOM. However, none of the other fluorescence indices (i.e., FI, E2/E3, and β/α) were significantly affected by prescribed fires, indicating that the source (mostly terrestrial) and molecular weight of DOM in leachate waters were mostly similar regardless of management.

3.5. Prescribed fire does not change the reactivity of DBP precursors

Average THM FP, HAA FP, and CH FP values were 61, 31, and 21 $\mu\text{g/L}$ for CW, respectively. DBP FPs for leachate waters were, however, significantly higher ($P < 0.05$) than for CW. Control water FP results were significantly lower ($P < 0.05$) than those of leachate water values. Thus, to evaluate the yield of DBP precursors from litter only, control water DBP FPs were subtracted from other DBP FP levels. Results showed that THM, HAA, and CH FPs of managed samples were significantly lower and mostly overlapped with each other with lower variability ($p > 0.05$) (average THM FPs = 2573 $\mu\text{g/L}$, 2223 $\mu\text{g/L}$, 2146 $\mu\text{g/L}$; HAA FPs = 2456 $\mu\text{g/L}$, 2325 $\mu\text{g/L}$, 2262 $\mu\text{g/L}$; and CH FPs = 365 $\mu\text{g/L}$, 271 $\mu\text{g/L}$, 273 $\mu\text{g/L}$ in pre-burned, dormant season and growing season burned samples, respectively) than unmanaged DBP FPs (THM FPs = 1143–7281 $\mu\text{g/L}$ with an average of 4354 $\mu\text{g/L}$; HAA FPs = 1531–7399 $\mu\text{g/L}$ with an average of 4108 $\mu\text{g/L}$; and CH FPs = 224–949 $\mu\text{g/L}$ with an average of 404 $\mu\text{g/L}$) (Fig. 4a, b, and c). Both THM and HAA FP concentrations were governed by DOC concentrations

of the samples over time, and have shown strong positive linear ($R^2 \geq 0.7$) correlations (Figure S11). However, CH FP vs. DOC correlations were weak for UW and GSW and strongly positive for MPW and DSW.

During the experiment, trichloromethane (TCM) constituted 98% of THMs (data not shown). Dichloroacetic (DCAA) and trichloroacetic acids (TCAA) constituted 30–35% and 61–66% (Figure S12a and b) of HAAs in all samples, regardless of treatment, and similar trends were found with total HAA FPs. This was related to limited Br⁻ presence in the samples. We also evaluated changes in DOC normalized yields ($\mu\text{g/mg-C}$) of DBPs. Results showed that the reactivity of THM (average 90–103 $\mu\text{g/mg-C}$), HAA (average 101–105 $\mu\text{g/mg-C}$), and CH (average 11–15 $\mu\text{g/mg-C}$) precursors did not change significantly over time, and have shown similar variations and overlap (Fig. 4d, e, and f). Furthermore, DOC normalized reactivity of DCAA (average 30–36 $\mu\text{g/mg-C}$) and TCAA (average 64–70 $\mu\text{g/mg-C}$) were also comparable and did not change significantly (Figure S12c and d). Our findings further supported two previous studies in which decreases in THM formation after prescribed burning were reported (Majidzadeh et al., 2019, 2015). Overall, these findings showed that prescribed fire and its application season did not change the nature and behavior of DBP precursors significantly under chlorination conditions regarding THM, HAA, and CH formation. As indicated before (Uzun et al., 2015; Yang et al., 2015), general water quality parameters such as DOC, TDN, DON, and Br⁻ are not good predictors (R^2 values for linear correlations <0.5) for NDMA FP under chloramination conditions. During the very first flush (Jan. 15, 2016), the NDMA FP value in CW was 14 ng/L. Significantly higher values were measured in unmanaged (274 ng/L) and managed (55–63 ng/L) leachate waters (Fig. 4g). As time progressed, NDMA FPs were comparable for each sample and were governed by CW derived precursors. The contribution of leachates was not significant ($p > 0.05$). These results indicated that (i) terrestrial-derived DOM do not contribute NDMA precursors; (ii) previously accumulated NDMA precursors released quickly from litter materials; and (iii) no more contribution occurred during the sample collection period due to isolated experimental conditions (no external anthropogenic pollution).

3.6. Prescribed fire does not affect leachate treatability

During the experiment, 23 L of leachate waters were collected on four dates (initial flushes; Jan. 15 and 23, dry period; Mar. 15, and first flush after dry period; May 31) for unmanaged and managed samples. Treatability tests (coagulation + flocculation + settling) were conducted as explained in SI with unfiltered leachate waters. Since DOC concentrations were different in each sample, we adjusted DOC concentrations with distilled deionized water (DDW) to establish similar organic carbon content between samples (target content was 4.0 mg/L) levels. Table 2 shows average water quality parameters after adjustment. This approach enabled

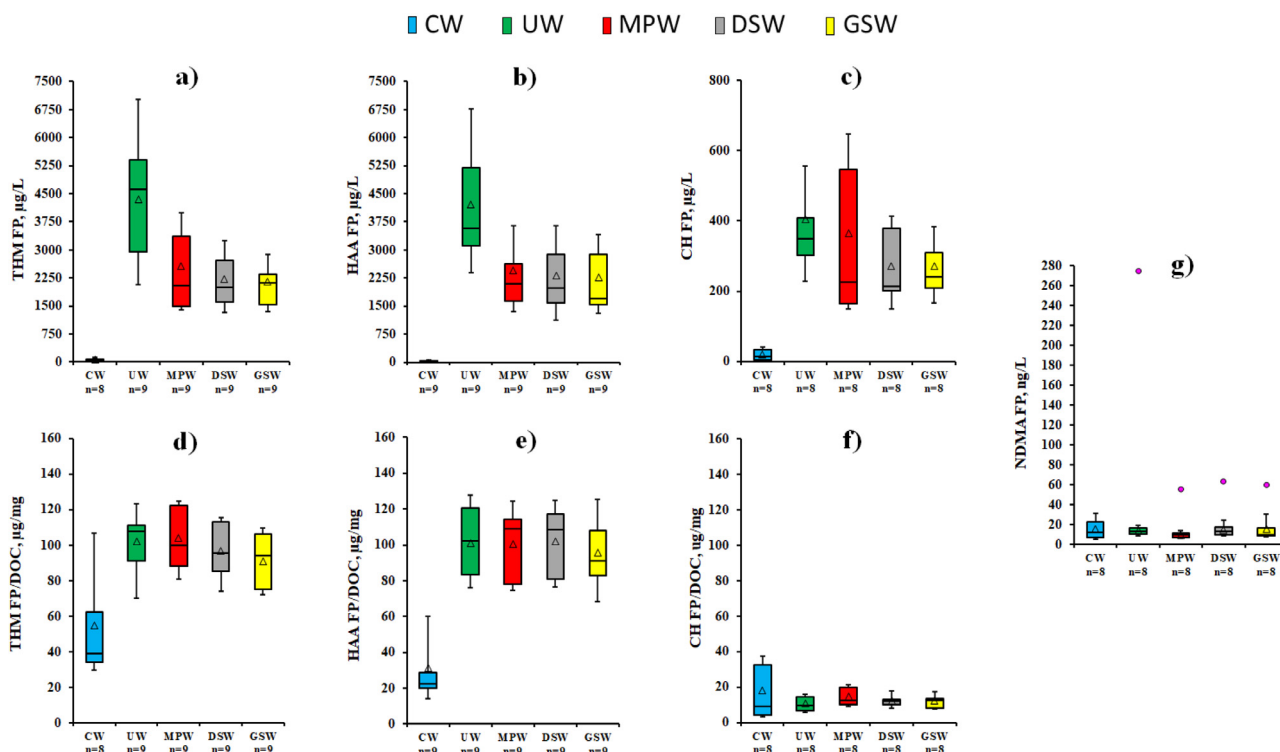


Fig. 4. FPs and carbon normalized FPs of DBPs of the leachate water for unmanaged and managed sites' leachate waters. THM FPs (a), HAA FPs (b), CH FPs (c), DOC normalized THM FPs (d), DOC normalized HAA FPs (e), DOC normalized CH FPs (f), and NDMA FP (chloramination only) (g). CW: Control water (rainwater), UW: Unmanaged leachate water, MPW: Managed pre-burned leachate water, DSW: After dormant season leachate water, GSW: After growing seasonal leachate water. n: indicates the number of data used for calculations. The box-plots used to represent data show 90th, 75th, 50th, 25th, and 10th quartiles, respectively. Triangles show the mean values of the data. Pink circles show outliers measured during the very first flush (Jan. 15, 2016).

Table 2

Comparison of diluted (DOC content adjusted) raw water quality parameters of leachate waters.

Samples (n = 4)	Water quality parameters				Optimum coagulant doses			
	DOC (mg/L)	Color (pt-co)	Turbidity (NTU)	SUVA ₂₅₄ (L/mg-m)	THM FP ($\mu\text{g/L}$)	HAA FP ($\mu\text{g/L}$)	^a Alum (mg/L)	^b Ferric (mg/L)
UW	4.4 ± 0.5	28 ± 10	1.6 ± 1.0	4.10 ± 0.79	441 ± 58	336 ± 185	30 ± 0	15 ± 0
MPW	4.0 ± 0.4	37 ± 27	1.9 ± 1.6	3.74 ± 1.05	379 ± 66	289 ± 155	30 ± 0	15 ± 0
DSW	4.2 ± 0.4	24 ± 13	1.4 ± 0.7	3.96 ± 1.23	399 ± 123	296 ± 225	30 ± 0	15 ± 0
GSW	4.2 ± 0.7	23 ± 2.0	1.3 ± 0.7	3.62 ± 0.94	384 ± 103	283 ± 216	30 ± 0	15 ± 0

^a Alum: aluminum sulfate eighteen hydrates ($\text{Al}_2[\text{SO}_4]_3 \cdot 18\text{H}_2\text{O}$).

^b Ferric: ferric chloride (FeCl_3). Errors show standard deviation between four samples. n: indicates the number of data used for calculations. UW: Unmanaged leachate water, MPW: Managed pre-burnt leachate water, DSW: After dormant season leachate water, GSW: After growing seasonal leachate water. Protein-like indicates tyrosine-like + tryptophan-like. Errors indicate standard deviation of six samples (n indicates the number of data used for calculations) measurement for each sample.

us to have similar coagulants to carbon ratio and to allow better comparisons in water treatability.

Results showed that coagulant demand (mg coagulant for mg removed DOC) for optimum DOC removals was not significantly affected by fire application. To reach optimal removal conditions, applied coagulant doses were similar for unmanaged and managed samples (alum = 30 mg/L and ferric = 15 mg/L), regardless of the burning season. The average DOC removal for unmanaged and managed pre-burned samples was similar ($p > 0.05$) (52% and 57% for alum and ferric, respectively). However, 5–10% lower DOC removals were observed in seasonally burned samples (Fig. 2a) compared to UW and DSW samples. This difference can be related to relatively lower fulvic acid-like content (Table 1) in these samples. Similarly, we evaluated the treatability of THM and HAA precursors in the same waters. THM FP removals were comparable for both coagulants (average removal was 68 ± 9% and 70 ± 9% for alum and

ferric, respectively). However, 5–10% higher removals were measured for HAA FPs compared to THM FP removals in leachate waters, regardless of burn season. Overall, results showed that prescribed burning did not cause significant changes in the treatability of DOC, THM, and HAA precursors.

4. Conclusion

A box model, Fig. 5 was created to summarize the effect of prescribed fire on i) the mass of forest floor materials, ii) water leaching of DOC, THM and HAA precursors from unburned vs. burned forest materials, and iii) treatability of those water leachates.

Our cumulative results indicated that low intensity, low severity prescribed fires may decrease fuel loads. This reduction induced only slight changes in DOM chemical functional groups associated with detritus materials. Continuous application of prescribed fires

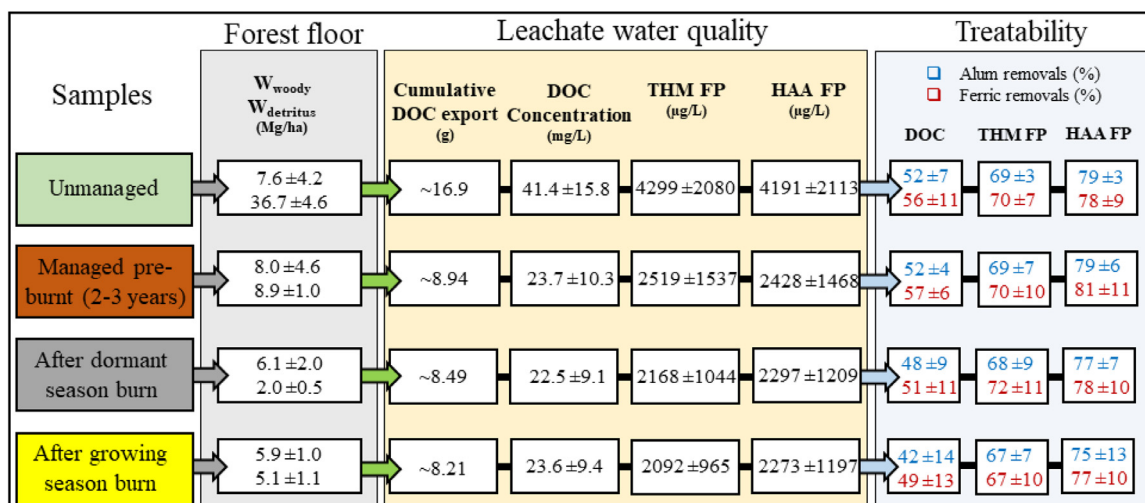


Fig. 5. The box model is summarizing the effect of prescribed fire on the forest floor, leachate water quality, and treatability. Errors show a standard deviation between three replicates. Cumulative DOC export (g) indicates total mass leached from 1 kg of mixture for ~1 year of the incubation period.

can decrease the leaching potential of DOC, TDN, DON, and reactive phosphorus from forest materials, thus lowering source water DOM concentration. The effect of prescribed fire may be limited to DOM composition, except for humic-like substances, which were reduced by fire. In addition to these potential benefits, prescribed fire is not expected to affect the release of Br^- and trace elements (K, Ca, Mg, Fe, S, Na, B, and Al) from forest materials. Thus, the formation of additional toxic brominated THMs and HAAs and the release of undesired elements are not of concern. Finally, periodically applied prescribed fire can reduce the export of THM and HAA precursors without changing carbon normalized yields ($\mu\text{g DBP FP/mg-C}$). Furthermore, the removal efficiency of DOC and precursors of THMs and HAAs under similar conditions (i.e., pH, organic carbon content, and coagulant dose) during alum or ferric coagulation is not expected to be changed after prescribed burning. Therefore, landowners can feel confident that low intensity, low severity prescribed fires do not negatively impact forest floor chemistry, water quality, or water treatability. The results would be further enhanced by conducting similar studies in different locations that contain different vegetation and site factors (i.e., hydrology, geology, soil type, etc.).

* Supporting Information

Detailed methods for water quality and DBP analysis, Table S1, and Figures S1–S12. Information is available free of charge on the XXX Publications website at the website at DOI: xxx.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.watres.2020.116385](https://doi.org/10.1016/j.watres.2020.116385).

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