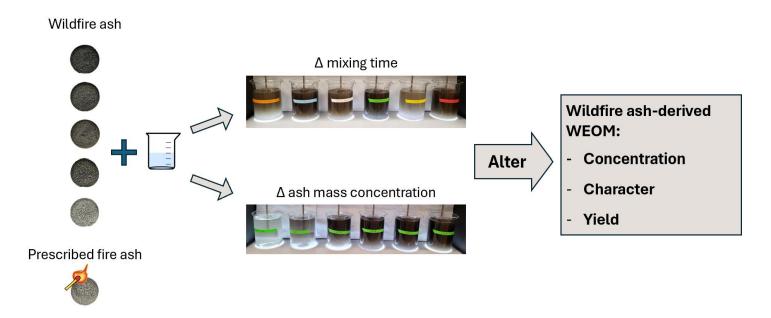
Methodological considerations for anticipating wildfire ash-associated organic carbon changes in water supplies

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Abstract

Wildfire can adversely impact the quality and quantity of water in forested regions by delivering excessive loads of sediment and burned materials into receiving waters via runoff. To evaluate implications to water treatability, bench- and pilot-scale investigations often involve wildfire ash addition to source water to reflect post-fire source water quality change. As methods are not standardized, a range of experimental conditions, such as different ash/water mixtures and mixing methods have been used—these can lead to contradictory results. Here, two key factors in source water preparation for investigating wildfire impacts on water treatability (i.e., mixing time and ash mass concentration) were investigated and their impacts on leached water extractable organic matter (WEOM) from wildfire ash were characterized. Specifically, a series of controlled bench-scale experiments were conducted to monitor and evaluate water quality changes in wildfire ash-impacted water (WAIW) at different mixing scenarios using both natural river water and ultrapure water. Water quality parameters including pH, alkalinity, conductivity, dissolved organic carbon (DOC), and specific ultraviolet absorbance (SUVA) were measured in WAIWs. Further characterization of organic matter involved liquid chromatography-organic carbon detection (LC-OCD) analysis. The concentration and character of organic carbon changed considerably during the first 24 hours of mixing. Water quality and ash mass concentration limit the extraction of organic matter into water. These results emphasize the importance of (i) specifying experimental conditions and providing rationale for the approach utilized (e.g., demonstrating a worst-case scenario of maximal leaching of WEOM from wildfire ash, reflecting watershed conditions), (ii) providing a range of results obtained at different leaching conditions, or alternatively (iii) clarifying that results may be exploratory or comparative, but not necessarily quantitatively meaningful or relevant for decision-making.

Graphical abstract



Keywords: 5-8

Climate change, treatability, water extractable organic matter (WEOM), water quality, drinking water, disinfection by-products (DBPs)

1 Introduction

Forested watersheds are critical sources of drinking water globally (UNFF, 2016; FAO, 2018). Anthropogenic and natural landscape disturbances, however, can deteriorate the high quality of water originating in these regions (Janetos et al., 1997; Christensen et al., 2004; Huntington et al., 2009; Whitehead et al., 2009; Watts et al., 2015). Although natural landscape disturbances are an integral part of ecosystem regulation, climate change has profoundly impacted the frequency and severity of such events (IPCC, 2018). Hotter and drier atmospheric conditions associated with climate change have contributed to the increased frequency of larger, more severe wildfires (USEPA, 2016). Wildfires can alter the timing of snowmelt and increase stream temperature (Wagner et al., 2014; Williams et al., 2019). They can also change the structure of soils and lead to decreased infiltration capacity and increased runoff (DeBano, 1991).

Higher rates of runoff can increase the transport and delivery of sediments, nutrients, and other contaminants from hillslopes to receiving waters (Bladon et al., 2008; Silins et al., 2009; Emelko et al., 2011). Warming temperatures, hydroclimatic changes, and disturbances such as wildfire also alter the mobility and delivery of organic carbon to water supplies (Emelko et al., 2024). The change in concentration, character and flux of dissolved and particulate organic carbon in rivers (Rhoades et al., 2019) can substantially challenge water treatment plant operations (Emelko & Sham, 2014; Hohner et al., 2016; Shams, 2018) and increase drinking water treatment costs (Emelko et al., 2011; Price et al., 2018). While nature-based

solutions such as biological filtration may offer some resilience to these shifts, their performance may also be compromised depending on organic carbon character (Blackburn et al., 2023).

Shifts in organic matter concentration and character have been reported following wildfires (Emelko et al., 2011; Hohner et al., 2016; Uzun et al., 2020, Tshering et al., 2023). The resulting changes in organic matter concentration and character can strongly affect drinking water treatment needs and performance. Ash collected from both burned landscapes following wildfires and those produced from simulated wildfires in a laboratory setting (Table 1) has been used in bench scale studies to evaluate water extractable organic matter (WEOM) concentration and character to inform wildfire impacts on water quality and treatability, including disinfection by-products (DBP) precursor reactivity.

To evaluate leaching of WEOM from wildfire ash, WEOM concentration and character have been evaluated using a range of experimental conditions (e.g., different ash-to-water ratios, and mixing conditions) (Table 1). However, the use of nonstandardized methods can often lead to contradictory results. For example, Wang et al. (2016) and Ahmed et al. (2023) modeled post-fire leaching of carbon during simulated rainfall events in laboratories, using two different approaches. Wang et al. (2016) attempted to replicate the cumulative impacts of rainfall on carbon leaching by subjecting a sample of pyrogenic materials to five sequential extractions with ultrapure water. Each time the samples were mixed for 2 hours, and the extracts were collected for further analysis. Ahmed et al. (2023) used three wet-dry scenarios on custom-made ash soil plots and passed deionized water through the plots for 15 minutes during each wetting event. Notably, these different leaching conditions resulted in contrasting trends in dissolved organic carbon (DOC) concentrations after the first simulated postfire rainfall event: Wang et al. (2016) observed a decreasing trend, while Ahmed et al. (2023) reported an increasing trend. Thus, the goal of this study was to investigate the potential change in organic carbon concentration and character using different mixing methods and identify practical mixing conditions that result in maximizing the concentration of WEOM from wildfire ash in water. Accordingly, the objectives of this research were to systematically evaluate (1) temporal changes of organic matter in wildfire ash-impacted waters (WAIW), and (2) the leaching potential of WEOM from wildfire ash into water at varying ash-to water ratios. The results provide guidance for preparing and characterizing WAIW to assess its treatability by identifying key factors affecting the extraction of organic matter from wildfire ash.

Table 1 Experimental conditions in previously reported studies on ash impacts on surface water organic matter concentration and quality. In studies where more than two ash-to-water ratios were used a range of ash mass concentration is presented.

Study	Type of ash/soil used & fire/burn severity	Mixing duration	Type of water used	Ash condition	Ash concentration (g/L)
Santin et al., 2012	wildfire ash- low to extreme	30 min shake & 20 min centrifugation	ultrapure	2.5 to 3 months post-fire	50
Olefeldt et at., 2013	wildfire burned soil	48 h stagnant & 1 h slow stirring	ultrapure	3 months post-fire	-
Audry et al., 2014	lab burned & wildfire ash- high	7 & 196 h & 66 days	deionized	fresh	2
Revchuk & Suffet, 2014	wildfire ash- low to high	48 h	ultrapure	fresh & 2 years post-fire	10
Wang et al., 2015a	wildfire ash- moderate & high	2 h	ultrapure	fresh	200
Wang et al., 2015b	Lab produced- moderate & high	2 h	ultrapure	-	20
Cerrato et al., 2016	lab burned- high	1, 24, & 72 h	ultrapure	-	3
Wang et al., 2016	wildfire ash- moderate & severe	5 times leaching	ultrapure	fresh	200
Cawley et al., 2017	wildfire affected sediment- low to high	2 min stirring & 6 or 24 h stagnant	river water	-	50
Hohner et al., 2017	wildfire affected sediment- low to high	2 min mixing & 24 h stagnant	river & low DOC tap water	-	13 to 64
Tsai et al., 2017	wildfire ash- moderate & high	72 h	ultrapure	fresh	250
Wilkerson, 2017	wildfire ash & soil- high	12 h & 15 min centrifugation	synthetic*	fresh	100
Cawley et al., 2018	lab burned soil- low to high	2 min stirring & 24 h stagnant	low DOC tap water	-	50
Rahman et al., 2018	lab burned- moderate to high	4, 24, 72 h	deionized	-	3.33
Tsai et al., 2019	wildfire ash- moderate & high	24 h	ultrapure	fresh	250
Chen et al., 2020	wildfire ash- moderate & high	24 h	rainwater	fresh & 1-year post-fire	1.35 to 52.7
Skwaruk et al., 2020	wildfire ash	2 min	river water	fresh	2 to 20
Thurman et al., 2020	lab burned- low to high	24 h	deionized	-	100
Chen et al., 2021	wildfire ash- moderate & high	24 h	ultrapure	fresh	250
Ferrer et al, 2021	wildfire, prescribed & lab burned	5 min	deionized	fresh & 1-year post-fire	50, 200
Wilkerson & Rosario-Ortiz, 2021	lab burned soil- low to high	6 h	ultrapure	-	5
Chen et al., 2022	lab burned- moderate to high	4 h	ultrapure	-	4
Rodela et al., 2022	wildfire ash- low to high	24 h	ultrapure	fresh	10
Wu et al., 2022	lab burned- moderate	48 h	reverse osmosis water	-	5
Ahmed et al., 2023	wildfire ash- very low to high	3 wet dry cycle (30 min wet)	deionized	fresh	-
Blackburn et al., 2023	wildfire ash- high	18 h & 3 h settling	high DOC river water	fresh	0.25 to 1
Chen et al., 2023	wildfire ash- moderate & high	24 h	rainwater	fresh & 1-year post-fire	1.35, 2.5
Dayaranthne et al., 2023	lab burned soil- low to high	24 h	ultrapure	-	0.25
Egan et al., 2023	lab burned litter & soil- unburned to high	3 days	synthetic*	-	20
Fischer et al., 2023	wildfire ash	16+ h	synthetic*	fresh & 1-year post-fire	-
Li et al., 2023	wildfire ash & lab burned- low to extreme	24 h stagnant & 10 min centrifugation	synthetic*	fresh	15
Muñiz González et al., 2023	wildfire ash- low to high	24 h shake	synthetic*	fresh	10
S´anchez-García et al., 2023	wildfire ash- low to high	5 min shake & 10 min settling	ultrapure	fresh & older	50
Thurman et al., 2023	wildfire ash- low to high	-	ultrapure	fresh	200
McGregor, 2024	wildfire ash - high	18 h mixing & 3 h settling	river water	fresh	1
Myers-Pigg et al., 2024	lab burned- low to high	24 h	synthetic*	-	25
Lersche et al., 2025	wildfire ash & soil	16 h agitation & centrifuge	synthetic*	fresh & 1-year post-fire	100
Numan et al., 2025	wildfire ash-very low to high	7 days	ultrapure	fresh	10
Roebuck Jr et al., 2025	lab burned- unburned to high	24 h	synthetic*	-	25

^{*} Synthetic refers to a variety of lab-produced water matrices.

2 Materials and Methods

2.1 Experimental approach

A bench scale study was conducted with controlled conditions to systematically evaluate the impact of (1) mixing conditions and time and (2) different ash-to-water ratios on the concentration and character of WEOM leached from wildfire ash. The six ash samples collected from different wildfires and one prescribed fire reflect a range of fire severities and ash characters. Synthetic WAIW matrices were prepared using high-quality source water collected from the Elbow River in Calgary, Alberta, Canada as well as ultrapure water. The use of ultrapure water allows for benchmarking of organic matter leaching from wildfire ash between sites in absence of water matrix effects while the use of natural source water is more system specific because it includes matrix effects.

Several studies focusing on the impacts of wildfire and/or lab-produced ash on water quality have used the terms "extraction" and "leaching" interchangeably to describe the transfer of organic matter from ash to water. While extraction procedures typically include testing conditions that maximize the extraction of a targeted element or pollutant from solid phase (e.g., Voegelin et al., 2003; Ran et al., 2007; Guigue et al., 2014), leaching tests usually intend to simulate the natural dissolution of compounds into water (e.g., Voegelin et al., 2003; Guigue et al., 2014). Thus, the release of organic matter from wildfire ash is described here as extraction, while the dissolution of organic matter into water is referred to as leaching, following the definitions provided in LEAF Methods 1313, 1315, and 1316 (EPA, 2023).

2.2 Experimental design

The overall design of the two bench scale experiments reported herein is presented in Figure 1 . The impacts of mixing conditions and time and ash mass concentration on WEOM were evaluated using Elbow River (Calgary, Alberta, Canada) source water and ultrapure (Milli-Q®, 18.2 M Ω -cm @ 25°C, TOC \leq 5 ppb) water. Elbow River water quality is summarized in Table 2. Notably, the temperature of WAIW matrices was maintained at approximately 21°C throughout the experiments.

Table 2 Elbow River water quality (n=8)

Parameter	Value
Turbidity (NTU)	1.00-5.00
рH	8.0-8.5
Alkalinity (mg/L as CaCO ₃)	140-170
SUVA (L/mg.m)	1.00-3.00
DOC (mg/L)	1.0-2.0

Experiment 1 involved two phases investigating the impact of: I) mixing conditions, and II) the duration of ash and water mixing on WEOM concentration and quality. In phase I, to evaluate the impact of mixing conditions 2 g of ash from two wildfires were mixed in 1 L of ultrapure water at two conditions: A) mixing for 5 minutes at 120 RPM followed by 24 h of settling; and B) mixing for 24 h at 120 RPM. Water samples were collected after 5 minutes of mixing and at the end of the experiment (i.e., after 24 h). The concentration of total and dissolved organic matter (i.e., TOC and DOC) were measured; dissolved organic matter was also characterized using specific ultraviolet absorbance at 254 nm (SUVA).

Based on the results from phase I of the experiment, mixing conditions for phase II were selected to maximize the concentration of WEOM from wildfire ash. Thus, synthetic WAIW matrices were prepared by mixing 2 g/L of each of six ash types with ultrapure and source water collected from Elbow River at 120 RPM for 24 h followed by 24 h of gentle mixing at 40 RPM (Figure 1). Subsequent samples were collected at the beginning of the experiment before ash addition to water (labeled as initial), and after the addition of ash when the suspension was mixed for 30 min, 6, 10, 24, and 48 hours. DOC, conductivity and pH were measured on all WAIW; dissolved organic matter was also characterized using SUVA and size exclusion liquid chromatography with organic carbon detection (LC-OCD) analyses. In addition, disinfection byproduct formation potential (DBPFP) and uniform formation condition (UFC) tests were performed on the WAIWs collected after 30 min and 24 h of mixing to demonstrate the mixing time impacts on the leaching of DBP precursors from ash to water. These analyses are further described in section 2.4.

In Experiment 2, the impact of ash mass concentration on WEOM concentration and character was evaluated. Two batches of synthetic WAIW with ash-to-water ratios of 0.5, 1, 2, 3, 4, and 5 g/L were made by mixing each of the six ash types with ultrapure and source water collected from the Elbow River in Calgary, Alberta, Canada (Figure 1). WAIW samples were collected after 24 h of mixing at 120 RPM, and their quality was characterized in the same manner as in the first set of experiments.

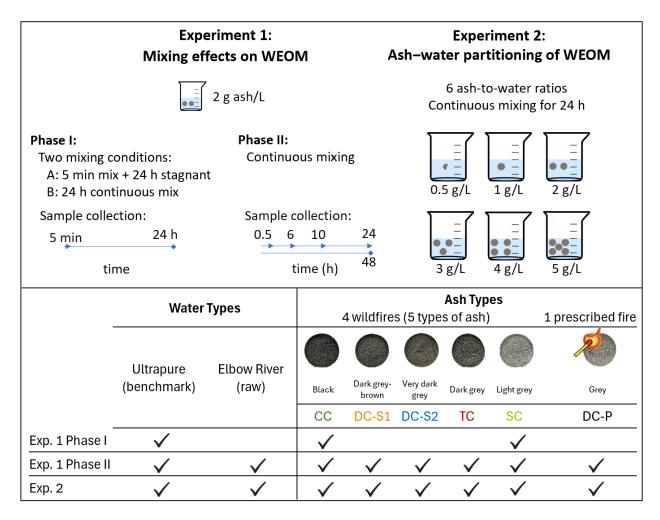


Figure 1 This study consisted of two experiments: Experiment 1 investigated the impact of mixing condition and time, while Experiment 2 examined the effect of ash mass concentration on the concentration and characteristics of WEOM. In each experiment, both ultrapure and natural untreated river water were used to prepare WAIW. Six different ash types were analyzed: CC (Cameron Creek ash), DC-S1 (Doctor Creek ash sampling site 1), DC-S2 (Doctor Creek ash sampling site 2), TC (Thuya Creek ash), SC (Scottie Creek ash), and DC-P (Doctor Creek ash after a prescribed fire).

2.3 Collection of wildfire ash

Ash samples collected from four wildfires and one prescribed fire reflect a range of pyrogenic materials produced by different burn severities (Table 3). All ash types were air dried and sieved through a 1 mm screen to homogenize samples and eliminate any large debris and conifer needles (Cawley et al., 2018; Harper et al., 2019; Skwaruk et al., 2020). The color of different ash types was identified visually using a Munsell color chart. Processed samples were stored in closed containers at a controlled temperature of 21°C. Organic matter content of each ash type was measured using loss on ignition according to ASTM D7348-21 method.

Table 3 Ash collection location, date and fire information, for the six ash types used in this study.

Ash name	Fire name	Location (Canada)	Burn area (ha)	Fire start date (mm/yyyy)	Vegetation type	Burn severity	Ash collection date (mm/yyyy)
Cameron Creek ash (CC)	Kenow Wildfire	Waterton, AB	35,000	09/2017	Lodgepole Pine1, Subalpine Fir ²	High to extreme	06/2018
Doctor Creek ash sampling site 1(DC-S1)	Doctor Creek Wildfire					High	
Doctor Creek ash sampling site 2 (DC-S2)	Doctor Creek Wildfire	Canal Flats, BC	7645	08/2020	Lodgepole Pine ¹ Douglas Fir ³ , Ponderosa Pine ⁴ , Western Larch ⁴	Medium	09/2020
Doctor Creek ash after prescribed fire (DC-P)	Doctor Creek Prescribed Fire					Medium	
Thuya Creek ash (TC)	Little Fort Fire Complex	Little Fort, BC	556	07/2017	Lodgepole Pine ¹ , Douglas Fir ³ , Spruce Hybrids ⁶ , Paper Birch ⁷	High	08/2017
Scottie Creek ash (SC)	Elephant Hill Fire Complex	Ashcroft, BC	200,000	07/2017	Douglas Fir ¹ , Spruce Hybrids ⁶ , Yellow Pine ⁸ , Trembling Aspen ⁹	High	08/2017
1- Pinus contorta var. latifa 2Abies lasiocarpa		6- <i>Ge</i>	rix occiden enus Picea		9- <i>Po</i> ₁	pulus tremui	loides

³⁻ Pseudotsuga menziesii var. glauca Pinus ponderosa 7- Betula papyrifera

⁴⁻ Pinus ponderosa

2.4 Water quality analyses

Most water quality analyses were conducted according to Standard Methods 24th edition (APHA, 2022). Water quality parameters investigated included pH (Standard Method 4500H using Fisher Scientific Accumet AB250 benchtop PH/ISE meter), alkalinity (titration method Standard Method 2320B using 0.1N HCl), conductivity (Malvern Panalytical Zetasizer Nano Z series with an accuracy of 10%) and maximum conductivity (200 mS/cm). DOC and TOC were measured using the combustion method (Standard Method 5310C) on a Shimadzu TOC-VCHP Analyzer (Shimadzu Corporation, Jiangsu, China) with a detection limit of 0.1 mg/L. The UV absorbance at 254 nm was measured using a UV₂₅₄ analyzer (RealTech P200B) following Standard Method 5910B. Specific ultraviolet absorbance at 254 nm (SUVA) was calculated by dividing UV₂₅₄ by DOC concentration (Edzwald et al., 1985; Weishaar et al., 2003).

Size exclusion liquid chromatography in combination with organic carbon detection (LC-OCD) analysis was conducted according to Huber et al. (2011) using a Model 8 LC-OCD analyzer (DOC-Labor GmbH, Karlsruhe, Germany). A size exclusion column, weak cation exchange column on a polymethacrylate basis (Toyopearl HW 50S, 250 mm x 20 mm, 30 µm from TOSOH Bioscience) was used for separation. The organic carbon detector was calibrated using potassium hydrogen phthalate. A customized software program (ChromCALC, DOC-LABOR, Karlsruhe, Germany) was used for data acquisition and processing. LC-OCD subdivides organic carbon into two general groups of hydrophobic (including all the molecules that retain on the column) and hydrophilic fractions, which can be further sub-divided into five fractions: biopolymers, humic substances, building blocks, LMW acids, and LMW neutrals based on molecular size.

Biopolymers are the largest group of hydrophilic compounds with molecular weights higher than 20,000 g/mol (>10 kDa) that don't absorb the 254nm UV radiation, and include polysaccharides, proteins, polypeptides and amino sugars. Humic substances include humic and fulvic acids with a molecular weight range from 400 to 1100 g/mol. Building blocks are defined as humic substance-like material of lower molecular weight (300-500 g/mol). LMW acids consist of small aliphatic organic acids with molecular weights lower than 350 g/mol). LMW neutrals include weakly charged hydrophilic or slightly hydrophobic compounds like alcohols, aldehydes, ketones, sugars and amino acids. WAIW samples were filtered through 0.45 μm polyethersulfone (PES), Millipore Express[®] PLUS; Merck Millipore, Burlington, MA, for DOC, UV₂₅₄ and LC-OCD analyses.

DBP formation potential (FP) tests under standard conditions (Methods 5710 B and 5710 D (APHA, 2022), as well as uniform formation conditions (UFC) (Summers, 1996) were performed on the ash-water matrices. DBPs (trihalomethanes (THMs) and haloacetic acids (HAAs)) precursors from the ash-water matrix and the potential of DBP formation in drinking water distribution systems after chlorination of ash impacted waters were evaluated. While the uniform formation conditions test predicts DBP formation within drinking

water distribution systems (Summers et al., 1996), the true FP test evaluates the reactivity of DBP precursors and allows for WEOM comparisons. For both DBP-FP and DBP-UFC tests, chlorine residuals after chlorination were measured according to colorimetric Method 4500-Cl G (APHA, 20122) using N, N-diethyl-p-phenylene diamine (DPD). Trihalomethanes (THMs) were analyzed using purge and trap extraction and gas chromatography with electron capture detection Method 501.1 (USEPA, 1979). Total trihalomethanes (TTHMs) concentration was calculated as the sum of four THM compounds (trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and tribromomethane (TBM)). Five haloacetic acids including trichloroacetic acid (TCAA), dichloroacetic acid (DCAA), monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), and dibromoacetic acid (DBAA)) were measured according to Method 552.3 (USEPA, 2003), a liquid-liquid extraction and gas chromatography with electron capture detection method. HAA5 was calculated as the sum of the concentrations of these five compounds.

3 Results and Discussion

3.1 Mixing effects on WEOM

3.1.1 Changes in WEOM concentration

The total and dissolved concentrations of extracted organic matter at different mixing conditions (Experiment 1 Phase I) are presented in Figure 2 a to d. TOC and DOC increased following ash addition to ultrapure water in all WAIWs. DOC concentrations increased more with continuous mixing relative to no mixing (i.e., stagnant conditions; Figure 2 a&b); this is likely due to enhanced particle interactions and increased surface area exposure, which facilitate more efficient leaching of WEOM.

DOC concentrations increased throughout the experimental period. In contrast, TOC concentrations decreased when WEOM was leached at stagnant conditions—TOC decreased by 93% and 22% in CC and SC WAIW, respectively (Figure 2 c&d). Such differences may be attributable to ash particle size distribution and particulate organic carbon density variabilities, which affect settling velocities. Accordingly, these findings underscore the importance of carefully considering how mixing conditions during bench- and pilot-scale investigations of WAIW either (i) connect to and reflect landscape conditions and processes relevant to wildfire ash impacts on water quality or (ii) maximize WEOM concentration so that treatment performance investigations can be conducted efficiently.

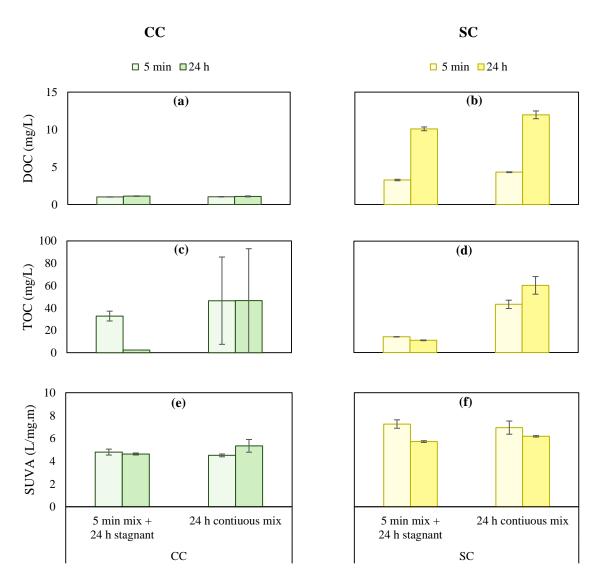


Figure 2 Change in a & b) DOC concentration (mean \pm SD, n=3), c & d) TOC concentration, and e & f) SUVA (mean \pm SD, n=3) for WAIW prepared using CC (Cameron Creek) and SC (Scottie Creek) ash mixed in ultrapure water at two different leaching conditions.

DOC concentration of WAIWs in Phase II are shown in Figure 3a. These concentrations ranged from 0.85 \pm 0.07 mg/L to 14.0 \pm 1.02 mg/L. Consistent with the results presented above and reported elsewhere (e.g., Rahman et al., 2018), the addition of wildfire ash to both ultrapure and Elbow River water resulted in an immediate (i.e., within the first 30 minutes of mixing) increase in WEOM concentration, this trend continued for 24 hours regardless of ash type. The changes in DOC concentration decreased during the second half (i.e., 24 to 48 h) of the experiment for all WAIWs (Table 4). The observed shifts in extracted organic carbon concentration after 24 h of mixing may be associated with photo/biodegradation (Bruun et al., 2008, Wagner et al., 2015); however, identification of a causal relationship was beyond the scope of the present investigation. Nonetheless, the substantial increase in DOC concentration, especially within the first

24 hours of the experiment, highlights the critical role of mixing time on the concentration of leached organic matter and consideration of the objective of the WAIW preparation (i.e., reflecting landscape process vs. creating a WAIW matrix for treatment performance or other evaluation).

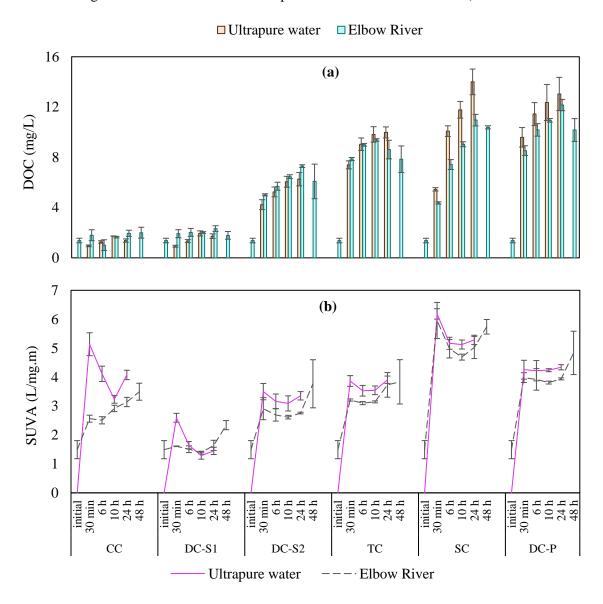


Figure 3 a) Change in DOC concentration, and b) SUVA. Light brown and turquoise bars show the changes in DOC concentrations for WAIWs prepared using ultrapure and Elbow River water, respectively. Solid purple and dashed grey lines show SUVA (mean \pm SD, n=3) in WAIWs prepared using ultrapure and Elbow River water, respectively. Initial sampling points represent values for ultrapure/raw water with no ash for both DOC and SUVA.

DOC concentrations increases during the first 24 h of mixing varied among ash types (Figure 3a) with SC and CC ash demonstrating the highest and lowest rates (12.1 and 8.3 mg/L DOC for SC ash and 0.5 and 0.2

mg/L for CC ash in ultrapure and Elbow River water, respectively). Variable DOC release from different wildfire ash types suggests that multiple factors such as burn severity, antecedent rainfall, and/or presence of soil mixed with the ash may substantially impact WEOM leaching—while such relationships are plausible and often suggested (e.g., Rahman et al., 2018; S'anchez-García et al., 2023), they can be confounded by differences in experimental approach as shown herein.

It is worth noting that a weak correlation between ash color and burn severity has been suggested (Bodi et al., 2011). Often, lighter colored (e.g., white or light grey) ash is attributed to more complete combustion Bodi et al., 2014; Rodela et al., 2022) and is thus expected to release less organic matter to water. It is also then suggested that darker-colored (e.g., black) ash will release more organic matter to water. In contrast, several studies have reported no appreciable differences in WEOM leached from white and black ash (Wang et al., 2015a; Tsai et al., 2017; Rodela et al., 2022), and in some cases, WEOM concentrations leached from white ash were substantially higher than those from black ash (Wang et al., 2016). Notably, in the present investigation, lighter colored ash types (i.e., SC and DC-P) released the highest concentrations of organic carbon to the water (Table 4).

Table 4 Change in DOC concentration in different WAIWs prepared using Elbow River water. The increases in DOC after 0.5 h and during the first (i.e., 0.5 to 24 h) and second (i.e., 24 to 48 h) half of the experiment are shown here.

Ash type	Initial increase in DOC within 30 min of the experiment ΔDOC _{0.5} (mg/L)	Change in DOC between 30 min to 24 h of experiment ΔDOC _{0.5-24} (mg/L	Change in DOC within the second 24 h of experiment ΔDOC ₂₄₋₄₈ (mg/L)	Ash color	Ash photo
CC	0.4± 0.3	0.2± 0.2	0.0± 0.2	Black	
DC-S1	0.5± 0.2	0.4± 0.1	-0.5± 0.2	Dark grey- brown	
DC-S2	3.6± 0.2	2.3± 0.1	-1.2± 1.5	Very dark grey	

TC	6.5± 0.3	0.8± 0.6	-0.8± 0.4	Dark grey	
SC	3.0± 0.2	6.6± 0.5	-0.6± 0.5	Light grey	
DC-P	7.2± 0.3	3.6± 0.1	-2.0± 1.3	Grey	

3.1.2 Changes in WEOM character: SUVA and LC-OCD

The changes in SUVA for CC and SC WAIW mixed at different conditions are presented in Figure 2 e&f. SUVA increased regardless of ash type and mixing condition (Figure 2 e&f). This observation was expected and aligns with previously reported studies (Tsai et al., 2019, Chen et al., 2020; Skwaruk et al., 2020). Similar to phase I, in phase II an initial increase in SUVA was recorded for all ash types, regardless of the source water quality (Figure 3b).

Longer ash-water contact time generally resulted in decreased SUVA in SC WAIW (Figure 2f). In contrast, while SUVA slightly decreased in CC WAIW at stagnant conditions, it increased with continuous mixing (Figure 2e). Changes in WAIW SUVA during mixing were also different between ash types in phase II (Figure 3), suggesting the potential for differential leaching dynamics between ash types.

Comparison of WAIWs prepared using ultrapure and Elbow River water shows that changes in SUVA values were more pronounced in WAIW matrices prepared with ultrapure water. Notably, SUVA was higher in WAIWs prepared using ultrapure water throughout the study period (Figure 3b). However, this difference became less noticeable for ash types with higher concentrations of leachable organic matter (i.e., SC and DC-P). This could be attributed to the effect of the natural organic matter (NOM) from Elbow River on SUVA of WAIW. The contribution of NOM from Elbow River water to the overall SUVA of WAIW was lower in WAIW with high WEOM/NOM ratios (SC and DC-P; Figure 3b).

Differential leaching dynamics associated with the molecular size and character are indicated by the LC-OCD analysis (Figure 4a). Here, the concentrations of refractory fractions (including the predominant fractions of WEOM: humic substances and building blocks) changed at different mixing times in the various WAIW (Figure 4). Notably while the yield of refractory fractions (i.e., sum of the concentrations of

refractory fractions divided by DOC concentration) generally remained stable during mixing, SUVA increased, decreased, and then increased further in all cases, reflecting the nonlinearity and complexity of leaching dynamics and underscoring that results from investigations reliant on prepared WAIWs must be interpreted cautiously (Figure 4b).

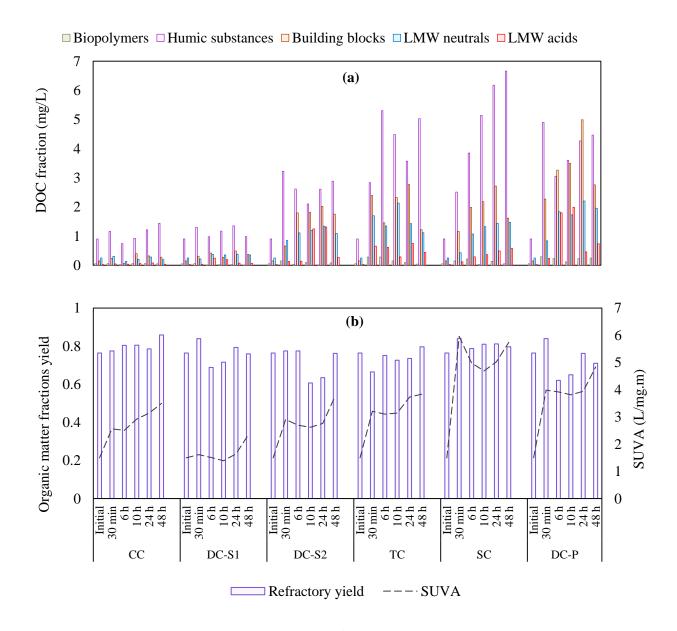


Figure 4 Change in organic matter character over time for WAIWs prepared in Elbow River water. a) Detailed representation of different fractions of organic matter. These fractions are presented in descending order of molecular weight: biopolymers, humic substances, building blocks, LMW acids and LMW neutrals; and b) Comparison between refractory fractions (including humic substances and building blocks) yield and SUVA.

3.1.3 Changes in WEOM extraction: Source water quality impact

In addition to the effects of source water NOM concentration and character on wildfire ash WEOM leaching, the potential impacts of source water pH, alkalinity, and conductivity on WAIW quality were also investigated. Figure 5 shows the pH of WAIW prepared using ultrapure and Elbow River. The changes in pH of the WAIWs prepared using Elbow River water after the addition of ash were negligible, likely due to the high alkalinity of Elbow River water (Table 2- Figure 5). In contrast, the absence of buffering capacity in ultrapure water resulted in a substantial increase in pH following ash addition. Additionally, the decrease in pH in the WAIW matrices prepared using ultrapure water towards the end of the 24 h experiment may be attributed to the increasing concentration of humic substances, and dissolution of atmospheric CO₂ into the WAIWs (Figure 5).

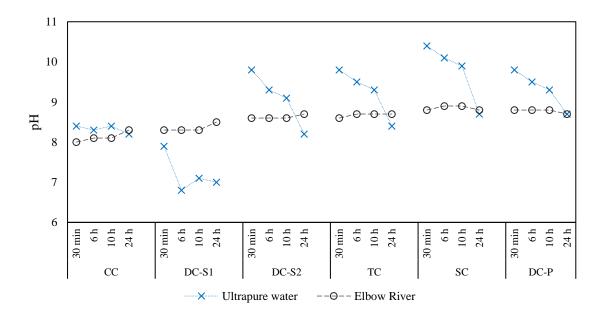


Figure 5 pH for all WAIWs prepared in ultrapure and Elbow River water during the first 24 h of experiment. Blue crosses and grey circles indicate pH of the WAIWs prepared in ultrapure and Elbow River water, respectively. The initial pH of Elbow River water was 8.3 ± 0.1 .

Table 5 presents conductivities of WAIWs prepared with 2 g/L of ash after 24 hours of mixing. Conductivities of WAIWs prepared using ultrapure water were consistently lower than those prepared with Elbow River water, primarily due to the higher initial conductivity in the Elbow River matrix. The net change in WAIW conductivity was consistently lower in the Elbow River matrix. While the impact of source water conductivity on organic matter extraction from wildfire ash has not been previously reported, an inverse relationship between source water conductivity and the dissolution of organic matter from soils

has been previously reported (Kalbitz et al., 2000; Säurich et al., 2017; Tiemeyer et al., 2017). Consequently, a greater change in DOC concentrations overtime was expected in WAIWs prepared with ultrapure water, relative to those prepared with Elbow River water (Figure 6).

Table 5 Conductivity (mean \pm SD) of the WAIWs made with 2 g/L of each ash type after 24 h of mixing. a) shows conductivities for WAIWs prepared in ultrapure water and b) provides conductivity values for those prepared in Elbow River water, as well as the average change conductivity from ash alone (calculated by subtracting conductivity of WAIWs from initial conductivity of Elbow River water).

(a)				
WAIW in ultrapure water	Conductivity (µS/cm)			
Ultrapure	0.00 ± 0.005			
CC	0.05 ± 0.002			
DC-S1	0.03 ± 0.001			
DC-S2	0.14 ± 0.03			
TC	0.13 ± 0.05			
SC	0.23 ± 0.06			
DC-P	0.21 ± 0.02			

(b)					
WAIW in Elbow River water	Conductivity (µS/cm)	Δ Conductivity (μS/cm)			
Elbow River	0.50 ± 0.03	-			
CC	0.51 ± 0.08	0.01			
DC-S1	0.51 ± 0.04	0.01			
DC-S2	0.61 ± 0.03	0.11			
TC	0.55 ± 0.04	0.05			
SC	0.66 ± 0.03	0.16			
DC-P	0.69 ± 0.05	0.19			

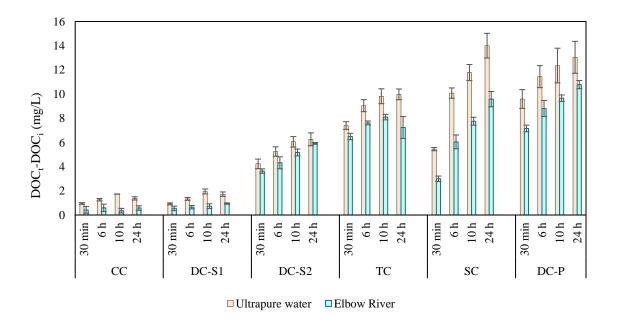


Figure 6 Change in DOC concentration (DOC at time t - source water initial DOC) and pH for all WAIWs during the first 24 h of experiment. Light brown and turquoise bars show the changes in DOC concentrations for WAIWs prepared ultrapure and Elbow River water, respectively. Changes in DOC concentrations were calculated by subtracting the DOC of WAIW at each sampling time (DOC_t) from the initial DOC of source water (DOC_i). Initial DOC concentration of ultrapure and Elbow River water was equal to zero and 1.47 ± 0.08 (mean \pm SD, n=6), respectively.

3.1.4 Changes in disinfection by-product precursors

DBP formation of WAIWs prepared using 2 g/L SC ash mixed in Elbow River water was measured after 30 min and 24 h of mixing. True formation potential (i.e., DBP-FP) and formation at uniform formation conditions (i.e., DBP-UFC) were evaluated. DBP precursor concentrations increased by approximately three times (i.e., from 3.99 ± 0.12 mg/L to 10.9 ± 0.40 mg/L; Figure 7) and led to a generally proportional increase in DBP formation, as would be expected. Notably, WAIWs mixed for 24 h had almost three times higher concentrations of DBPs compared to those that were mixed for 30 min, irrespective of DBP type (i.e., THM or HAA) and chlorination conditions (FP or UFC; Figure 7). Shorter reaction times and lower chlorine doses at UFC condition (Summers et al., 1996; APHA, 2022) resulted in generally lower DBP-UFC compared to DBP-FP (Figure 7 7). This aligns with what has been reported previously (Yang et al., 2005).

Collectively, the results presented herein demonstrate that mixing time can substantially affect DBP precursor concentration and character and the concentration of DBPs produced during chlorination of WAIW. Accordingly, it is critical that evaluations of *prepared* WAIW matrices (as opposed to wildfire-impacted source waters) explicitly outline experimental conditions and justify the selected methodology, present a range of outcomes across various leaching conditions, or acknowledge that findings may not be quantitatively meaningful for decision-making and might be exploratory or comparative.

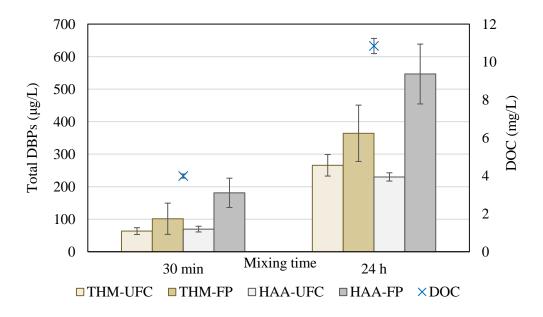


Figure 7 Change in DOC and DBP concentration (mean ± SD, n=3) in WAIWs prepared using SC (Scottie Creek) ash after 30 minutes and 24 hours of mixing in Elbow River water. Brown and grey bars represent concentrations of THMs and HAAs, respectively and blue crosses indicate DOC concentration at each mixing time.

3.2 Ash–water partitioning of WEOM

3.2.1 Changes in WEOM concentration

Figure 8 a&b shows variations in leached WEOM in WAIWs (i.e., DOC concentration) after 24 h of mixing with ash-to-water ratios ranging from 0.5 to 5 g/L. Notably, the lowest ash mass concentration (i.e., 0.5 g/L) resulted in considerable increase in DOC concentration (e.g., SC and DC-P; Figure 8 a&b). Suggesting ash characteristics can substantially impact the extraction of organic matter from wildfire ash. This aligns with previously reported studies (e.g., Rahman et al., 2018; S'anchez-García et al., 2023). Additionally, a positive correlation between ash-to-water ration and DOC concentration was observed over the ash mass concentrations investigated (i.e., WEOM saturation was not reached). For the same ash mass concentration, the highest WEOM concentration was measured in WAIWs prepared using SC and DC-P ash types, while it was considerably lower in WAIWs prepared with CC and DC-S1 ash, regardless of the water used (i.e., ultrapure or Elbow River; Figure 8 a&b). Although this observation did not align with the color-based classification of ash and its WEOM content, it was consistent with the findings of the previous experiment (section 3.1).

Ash organic matter content and WEOM concentration for the investigated ash types are presented in Table 6. Organic matter content ranged from $3.7 \pm 0.1\%$ to $21 \pm 1.9\%$ for DC-S1 and CC, respectively (Table 6). Notably, the lowest DOC concentrations were measure in WAIWs prepared using the same ash types. This suggests that there is no correlation between ash organic matter content and its leachable organic carbon concentration.

At ash mass concentrations ranging from 0.5 to 2 g/L, the normalized concentration of extracted organic matter (expressed as mg of DOC extracted per g of ash) was highly sensitive to ash-to-water ratio. WEOM yield decreased with an increase in ash mass concentration in all WAIW (Figure 8 c&d). For ash-to-water ratios exceeding 2 g/L, the normalized WEOM concentration stabilized, showing minimal further change, however. This behavior might be attributed to changes in ionic strength following the addition of ash to water (e.g., Brito et al., 2021), which can influence the solubility of organic matter (Lawrence & Roy, 2021). Specifically, this observation can be attributed to dissolution chemistry differences between dilute and non-dilute systems (Logan, 2012; Klučáková & Věžníková, 2016). In concentrated solutions and suspensions, ionic strength increases and electrostatic interactions become stronger, leading to deviations from ideality. The reasons for these deviations at higher ionic strength may include increased (i) competition between inorganic ions and organic molecules for water molecules that reduce the hydration shell around organic molecules rendering them less soluble and more likely to aggregate or precipitate (i.e., "salting out"), (ii) compression of the electric double layer around negatively charged organic matter

leading to its destabilization and aggregation thereby preventing or reducing its dissolution, (iii) cation bridging between negatively charged organic functional groups reducing solubility and promoting the formation of organic complexes (flocs) that limit dissolution, and (iv) reduced hydrophobic dissolution as a result of increased solution/suspension polarity at higher ionic strength making it less favorable for non-polar, hydrophobic organic molecules (e.g., aromatic and lipid-like components of DOC) to dissolve. Similar trends have been observed in studies examining the leaching of various inorganic elements from soils and fly ash. These studies consistently reported a decrease in leaching efficiency for multiple metals as the solids-to-liquid (S/L) ratio increases, highlighting the broader influence of solid concentration on solute mobility (Koralegedara et al., 2017; Cao et al., 2018; Ghosh & Kartha, 2025). The increase in S/L ratio reduces the available free water molecules to interact with solute particles, thereby hindering the solubility of various compounds, ultimately decreasing extraction efficiency.

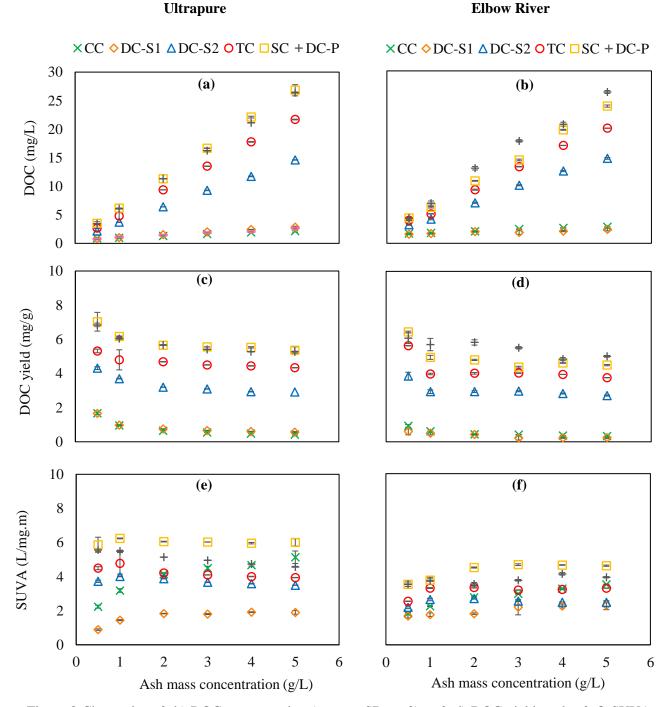


Figure 8 Change in a & b) DOC concentration (mean \pm SD, n=3), c & d) DOC yield, and e & f) SUVA (mean \pm SD, n=3) for different WAIWs prepared using ultrapure and Elbow River water across a range of ash-to-water ratios.

Table 6 Organic matter content, DOC concentration in WAIW containing 2 g/L of ash mixed in ultrapure water. The table is organized in descending order of organic matter content for different ash types.

Ash type	Organic matter content (Moist Free % wt)	Moisture Content (%wt)	DOC (mg/L)
CC	21.0 ± 1.9	3.9 ± 0.1	1.3 ± 0.04
DC-P	20.7 ± 0.1	1.6 ± 0.03	11.9 ± 1.0
DC-S2	17.0 ± 0.5	4.2 ± 0.01	6.3 ± 0.1
TC	14.7 ± 0.7	5.0 ± 0.02	9.6 ± 0.3
SC	14.6 ± 0.4	4.6 ± 0.1	12.2 ± 1.6
DC-S1	3.7 ± 0.1	3.9 ± 0.04	1.6 ± 0.1

3.2.2 Changes in WEOM character: SUVA and LC-OCD

Figure 8 e&f presents the changes in SUVA for WAIWs prepared using different ash mass concentrations. Regardless of the ash type, SUVA values increased WAIW at lower ash-to-water ratios (i.e., 0.5 to 2 g/L). However at ash mass concentrations greater than 2 g/L SUVA stabilized for almost all ash types, except for CC (Figure 8 e&f). SUVA values were higher in WAIWs prepared using ultrapure water, for almost all ash types (except for DC-S1), this could be attributed to the initial SUVA in Elbow River water and its potential impact on the overall WAIW SUVA.

LC-OCD analysis confirmed changes in WEOM character with increasing ash mass concentration, as shown in Figure 9a. The data shows that increasing ash-to-water ratio leads to an increase in the concentration of all organic matter fractions. Consistent with the results of experiment 1 (section 3.1) and previously reported studies (Skwaruk et al., 2020; Blackburn et al., 2023), the greatest increases in organic matter fractions were in refractory fractions of DOC (i.e., humic substances and building blocks). However, there is no apparent proportionality among increases of different fractions at each ash mass concentration (Figure 9a). In contrast to SUVA, refractory fractions yield generally remained stable in all WAIWs across different ash-to-water ratios. This discrepancy emphasizes that different methods of analysis may capture distinct aspects of the organic matter present and highlights the critical need for careful interpretation of results.

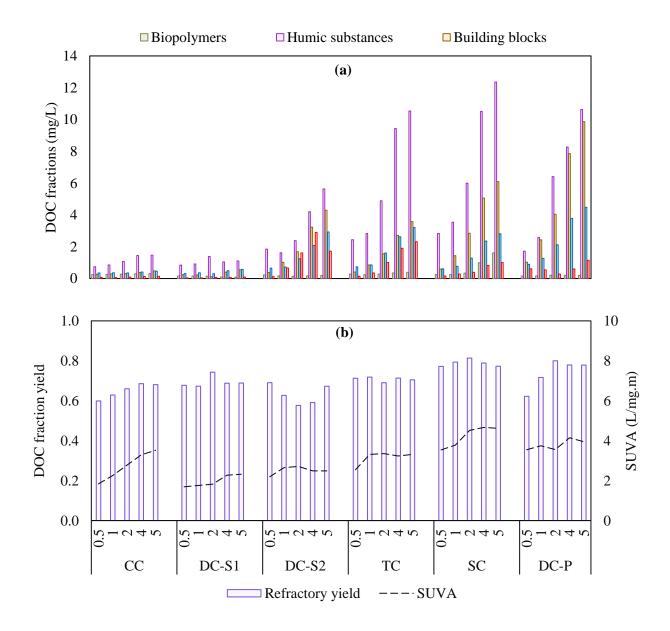


Figure 9 a) Change in organic matter character over time as indicated by LC-OCD for different ash types prepared using Elbow River water. a) Detailed representation of different fractions of organic matter. These fractions are presented in descending order of molecular weight: biopolymers, humic substances, building blocks, LMW neutrals and LMW acids; and b) Comparison between refractory fractions (including humic substances and building blocks) yield mg refractory fraction divided by mg DOC) and SUVA

3.2.3 Changes in WEOM extraction: Source water quality impact

The impacts of initial pH and alkalinity of source water on the overall WAIW pH were also assessed. Figure 10 shows changes in pH for all WAIWs prepared using different ash mass concentrations mixed in ultrapure and Elbow River water. The largest changes in pH were observed in WAIWs prepared using ultrapure

water. In contrast, changes in pH in Elbow River water were negligible, due to its high initial alkalinity (140-170 mg/L; Table 2).

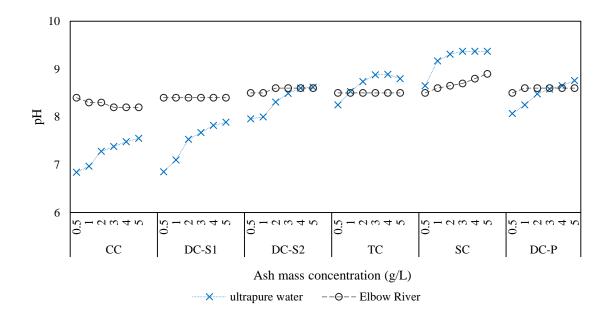


Figure 10 Change in pH for all ash types mixed in ultrapure and Elbow River water for different ash-to-water ratios. Blue crosses and grey circles indicate pH of WAIWs prepared in ultrapure and Elbow River water, respectively. The initial pH of Elbow River water was 8.3 ± 0.1 .

4 Conclusions

The effects of mixing conditions, time, and ash mass concentration on water extractable organic matter (WEOM) release from wildfire ash to water were investigated to demonstrate that experimental approach affects water quality and treatability assessments when using prepared wildfire ash-impacted water (WAIW) matrices (i.e., water matrices to which pyrogenic material is added to infer wildfire implications to water quality or treatability). Key conclusions from this research are:

- the impacts of wildfire on WEOM characteristics were generally consistent across different ash types. A higher DOC concentration consisting of mainly humic substances was measured in all WAIWs, regardless of mixing condition and ash mass concentration in the suspension,
- WEOM concentrations in ash leachates and ash organic content do not necessarily correlate with the ash color classification scheme,
- substantial changes in organic matter concentration and character occur during the first 24 h of WAIW mixing and can impact inferred implications of wildfire ash on drinking water quality and treatability,

- source water quality and concentration of ash added to prepare WAIW impact WEOM solubility and therefore inferred implications of wildfire ash on drinking water quality and treatability, and
- in absence of conducting a system-specific characterization of WEOM leaching from wildfire ash with different mixing conditions (or developing an alternative leaching method), a 24-hour mixing time is recommended for WAIW preparation for investigations (e.g., treatment performance evaluations) focused on maximizing WEOM concentration.

The conclusions above highlight several important considerations for conducting investigations of wildfire impacts on water quality and treatability in situations where wildfire impacts cannot be directly investigated. Most notably, they collectively emphasize the importance of (i) specifying experimental conditions and providing rationale for the WAIW preparation approach utilized (e.g., demonstrating a worst-case scenario of maximal leaching of WEOM from wildfire ash, reflecting watershed conditions), (ii) providing a range of results obtained at different leaching conditions, or alternatively (iii) clarifying that results may be exploratory or comparative, but not necessarily quantitatively meaningful or relevant for decision-making.

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Declaration of Competing Interest

The authors declare no competing financial interest.

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