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Two years of post-wildfire impacts on dissolved organic matter, nitrogen, and precursors of disinfection by-products in California stream waters

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ABSTRACT

We investigated the effects of two California wildfires (Rocky and Wragg Fires, 2015) compared to an unburned reference watershed on water quality, dissolved organic matter (DOM), and precursors of disinfection by-products (DBPs) for two years' post-fire. The two burned watersheds both experienced wildfires but differed in the proportion of burned watershed areas. Burned watersheds showed rapid water quality degradation from elevated levels of turbidity, color, and suspended solids, with greater degradation in the more extensively burned watershed. During the first year's initial flushes, concentrations of dissolved organic carbon (DOC), dissolved organic nitrogen (DON), ammonium (NH_4^+/NH_3), and specific ultraviolet absorbance ($SUVA_{254}$) were significantly higher (67 ± 40%, 418 ± 125%, 192 ± 120%, and 31 ± 17%, respectively) in the more extensively burned watershed compared to the reference watershed. These elevated values gradually declined and finally returned to levels like the reference watershed in the second year. Nitrate concentrations were near detection limits (0.01 mg-N/L) in the first year but showed a large increase in fire-impacted streams during the second rainy season, possibly due to delayed nitrification. Changes in DOM composition, especially during the initial storm events, indicated that fires can attenuate humic-like and soluble microbial by-product-like (SMP) DOM while increasing the proportion of fulvic-like, tryptophan-like, and tyrosine-like compounds. Elevated bromide (Br^-) concentrations (up to 8.7 μM]) caused a shift in speciation of trihalomethanes (THMs) and haloacetic acids (HAAs) to brominated species for extended periods (up to 2 years). Wildfire also resulted in elevated concentrations of N-nitrosodimethylamine (NDMA) precursors. Such changes in THM, HAA, and NDMA precursors following wildfires pose a potential treatability challenge for drinking water treatment, but the effects are relatively short-term (≤1 year).

1. Introduction

Forested watersheds are a principal source of drinking water around the world (Costanza et al., 1997). Considering the United States (US) as an example, more than 60% of potable water is sourced from forested areas (Smith et al., 2011; Stein et al., 2005). With rapid population growth and a changing climate, these vital water sources are vulnerable to both natural and human-made disturbances. For example, forest fires are of particular concern because they rapidly consume biomass, convert forest vegetation into pyrogenic organic matter (PyOM) and change the physical and chemical properties of upper soil layers (Goforth et al., 2005; Oliveira-Filho et al., 2018; Santín et al., 2015; Vergnoux et al., 2011; Wang et al., 2016). Consequently, surface run-off regimes that strongly affect the quantity and quality of water exported from forested catchments are markedly altered (Ellis et al., 2004; Knoepp et al., 2008). Furthermore, forested watersheds in arid and semi-

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arid regions worldwide are expected to experience higher temperatures and extended droughts, resulting in more frequent, intense, and massive wildfires (Bowman et al., 2009; Mehran et al., 2017; Smith et al., 2011; Williams et al., 2019).

PyOM materials create hydrophobic soil properties and foster the formation of surface crust, resulting in increased water repellency and reduced infiltration into soil profiles (Goforth et al., 2005). As a result, increased water yields or even flash floods/debris flows commonly originate from burned watersheds during post-fire rainstorms (Mahat et al., 2016; Malvar et al., 2015). Further, bare soil conditions accelerate soil erosion, transporting sediments and nutrients to source waters (Belillas et al., 1993; Lane et al., 2008; Malmon et al., 2007; Sheridan et al., 2007; Stevens, 2013). Additionally, elevated levels of turbidity and suspended solids (Hohner et al., 2017; Mast et al., 2016; Murphy et al., 2012) and metals (Abraham et al., 2017) are frequently reported in the post-wildfire runoff.

Fire temperature and oxygen availability during a wildfire are the principal factors affecting the consumption of detrital biomass and the formation of PyOM in wildfire ash. Thus, fire characteristics have a substantial effect on the quality and quantity of terrestrially-derived dissolved organic matter (DOM) exported from burned watersheds (Hohner et al., 2016; Wang et al., 2015, 2016). Higher fire temperature is expected to increase detritus pyro-mineralization (Gray and Dighton, 2006). Therefore, the amount of water leachable DOM, expressed as dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) concentrations, might be expected to be lower in PyOM than in the unburned organic matter from which it originated (Wang et al., 2016). However, higher DOC and DON concentrations are frequently reported in stream waters following wildfires (Hohner et al., 2016; Revchuk and Suffet, 2014; Writer et al., 2014). Such discrepancies can be related to enhanced mobility and flushing of DOM, especially during intense post-wildfire precipitation events (Bladon et al., 2008; Emelko et al., 2011; Hohner et al., 2016; Inamdar and Mitchell, 2007; Murphy et al., 2015; Stevens, 2013).

In terms of health-related adverse effects from wildfires, the occurrence of disinfection by-products (DBPs) is one of the main concerns in treated drinking water. Carbonaceous (C-) DBPs, trihalomethanes (THMs), and haloacetic acids (HAAs) are regulated in drinking water in the US (USEPA, 2006). However, recent studies have demonstrated that commonly applied secondary disinfection practices, such as chlorination, are shifting to chloramination (Li, 2011) which is related to the formation of nitrogenous N-nitrosodimethylamine (NDMA), a DBP with more significant health risks than those associated with regulated THMs and HAAs (Bond et al., 2011; Plewa et al., 2002; Richardson et al., 2007). Since fires have the potential to change the chemical characteristics and release of DOM from forest materials, the reactivity of DBP precursors originating from wildfires is expected to differ in stream runoff after wildfire events (Hohner et al., 2016, 2017; Revchuk and Suffet, 2014; Writer et al., 2014). Previous studies showed post-fire increases in DOC leaching from burned areas that can augment the levels of precursors for THM (Revchuk and Suffet, 2014) and HAA (Writer et al., 2014) precursor levels. Following post-wildfire rainstorms, nitrogenous (N-) DBP precursors, including NDMA, were high (Cawley et al., 2016; Hohner et al., 2016; Wang et al., 2015).

While previous studies demonstrated changes in DOM and various water quality parameters after wildfires, most of these studies were either conducted under controlled laboratory conditions or based on stream water sampling campaigns that did not fully achieve their purpose due to environmental and logistical restrictions. Currently, no study has systematically evaluated the first and second-year responses of different watershed conditions (i.e., headwater vs. higher-order and complete vs. partially burned watersheds) on post-wildfire runoff and their implications for DOM release and reactivity of DBP precursors. In this study, we collected water samples from wildfire impacted watersheds in northern California during all major post-fire runoff events covering two consecutive rainy seasons (2016 and 2017). We captured short- (first-year response including initial and subsequent flushes) and medium-term (second-year response) effects on stream water from two separate burned watersheds and one reference (i.e., unburned) watershed. The specific objectives of this study were to evaluate the temporal changes in post-fire surface water quality with a focus on changes in DOM release, characterization, and reactivity as DBP precursors in burned vs. unburned watersheds. The results of this study provide valuable information to drinking water providers for assessing potential risks that wildfires pose to drinking water safety.

2. Materials and methods

2.1. Wildfires and site description

In July–August 2015, the Rocky Fire, Jerusalem Fire, and Wragg Fire in the Northern California Coastal Ranges consumed 281 km^{2,} 102 km^2 and 33 km^2 of wildlands, respectively (Fig. 1). The Rocky and Jerusalem Fires started as two separate fires but eventually merged into a single fire. Thus, to avoid confusion, we referred them collectively as the Rocky Fire. Geographic information system (GIS) calculations and site observations indicated that the burn area affecting sampling stream was ~20% of Upper Cache Watershed (hydrological unit code 8 [HUC] – 18020116 but exclude the downstream area of sampling point), and >90% of the second-order watershed (a small portion of the HUC12–180201620503 McCune Creek-Putah Creek). In addition, based on a previous study (Wang et al., 2020) and the visual indicators (i.e., consumed duff, generally dark-colored ash, and blackened/dead standing trees) (United States Department of Agriculture [USDA], 2020), these fires can be characterized as medium severity. Pre-burn vegetation was a mixture of oak savanna and woodlands, including blue oak (*Quercus douglasii)*, interior live oak (*Quercus wislizenii*), and scrub oak (*Quercus dumosa)*; chaparral, including buckbrush (*Ceanothus cuneatus*), chamise (*Adenostoma fasciculatum*), California buckeye (*Aesculus californica)*, toyon (*Heteromeles arbutifolia*), and manzanita (*Arctostaphylos spec.*); and annual grasslands. Dominant soils were Inceptisols, and Alfisols formed on mixed sedimentary parent materials. Watersheds were dominated by moderate (15%) to steeply (45%) sloping topography resulting in high water erosion potential. The area has a Mediterranean climate, characterized by hot, dry summers and mild, rainy winters, with the majority of the precipitation occurring between November and March. Records for the Berryessa Dam (Fig. S1), located adjacent to the Cold Creek watershed, indicated that the area received ~431 and ~1029 mm of total precipitation during the first rainy season (Jan. 5 and April 10, 2016) and the second rainy season (Dec. 15, 2016, and March 31, 2017) (intense rainy seasons), respectively (Department of Water Resources [DWR], 2017). Rainfall patterns were identical between the reference and Wragg Fire watershed as they were located within a few km of each other. Winter rainfall events in northern California are large storms originating in the Pacific Ocean that affect large land areas. Unlike classic thunderstorms, these storms do not show large differences in precipitation at small spatial scales. Thus, we assumed that all three watersheds in this study received rainfall from the same storms, although the total rainfall amounts might not differ significantly among watersheds.

2.2. Water sample collection

Three sampling locations (outlets of watersheds) (Fig. 1) were

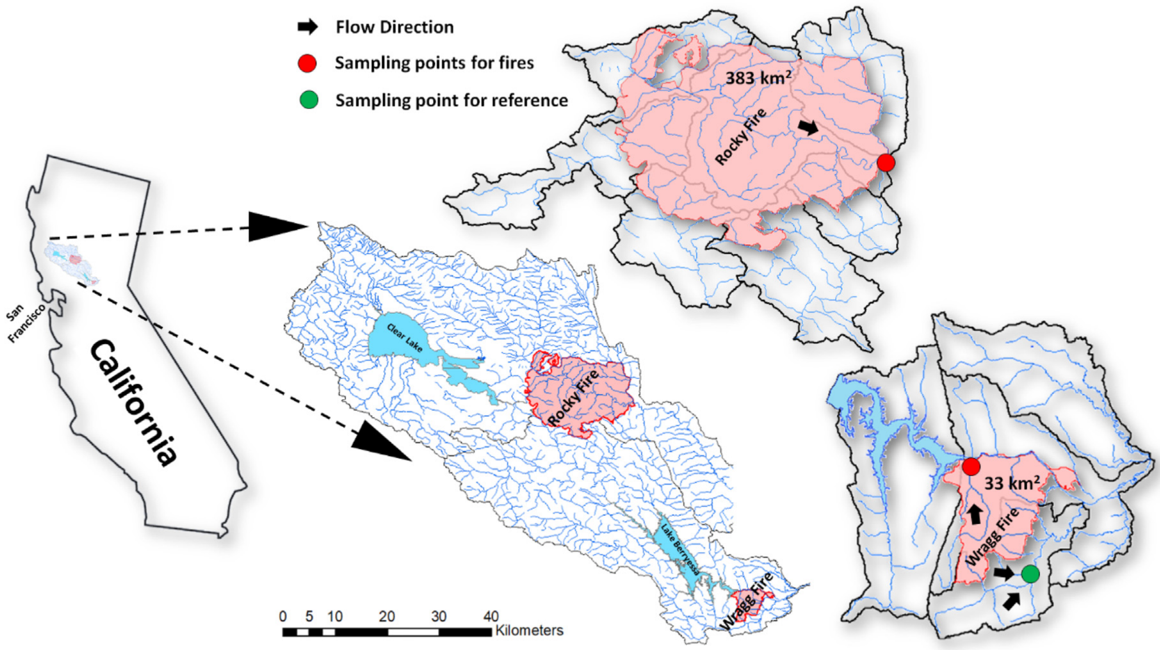

Fig. 1. The map shows the locations of the Rocky and Wragg Fires in Northern California. Red zones are the perimeters of the wildfires within watersheds. The Rocky Fire and Wragg Fire consumed a total of 383 km^2 and 33 km^2 wildlands, respectively. Three sampling locations were established to assess the effects of fires on water quality at the watershed scale. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

established to assess the effects postfire rainstorms on water quality at the watershed scale. Samples from the Wragg Fire were collected from the outlet of Cold Creek (38°30′43.31″N; 122° 5′50.02″W), which drains the significant part of a burned watershed. The quality of water samples collected from the burned watershed was compared to the quality of water samples collected from an adjacent reference watershed that was not affected by wildfire (Miller Canyon Creek, 8°27′4.94″N; 122° 3′22.97″W). The two watersheds have similar geology, soils, topography, and vegetation. Importantly, the Wragg Fire did not burn any of the land areas within the “reference” watershed. Therefore, the reference watershed served as a burned vs. unburned watershed comparison for the Wragg Fire. Streamflow from the Cold Creek and Miller Canyon Creek was intermittent with flow only during the rainy season. Thus, the sampling frequency was governed by weather conditions (Fig. S1). During the first year’s sampling, there were six, sixteen and seventeen reference, Rocky and Wragg Fire samples collected across the sampling period, respectively. But no reference sample for the very first flushes due to a hydrologic lag that resulted from the vegetated landscape. Nonetheless, we didn’t miss the collection of the early reference samples, it was a function of the hydrologic lag in generating streamflow. During the second year’s sampling, there were sixteen samples for reference and Rocky Fire and seventeen samples for Wragg Fire.

Regarding flow regimes, there were two distinct rainfall periods in the first rainy season (Jan. 5 and April 10, 2016) because of the lack of rainfall during February 2016. We defined the runoff from these two periods as “Season 1—initial flushes (S1—if)” (Jan. 5—22, 2016) and “Season 1—subsequent flushes (S1—sf)” (Mar. 5—Apr 10, 2016). In the second rainy season (Dec. 15, 2016—Mar. 31, 2017), rainfall was more continuous after the onset of the rainy season, and we labeled this period as “Season 2—flushes (S2—f)” (Fig. S1). For the Wragg Fire flows, we measured instantaneous discharge from the water velocity × cross-sectional area when the flows were below 0.14 m^3/s. For the flow >0.14 m^3/s, we determined the flow as the difference between Putah Creek flow (about 1.6 km downstream of Wragg input) (USGS station 11454000) minus dam release (Berryessa) from immediately upstream of the Wragg fire stream. The calculations were acceptable until Feb. 16, 2017 when the spillway overflowed and there were no reliable measurements for water releases from Berryessa Reservoir (Fig. S2a).

In terms of the geology and soil characteristics, the reference watershed (78 km^2) does not represent the larger Cache Creek watershed (3017 km^2) in which the Rocky Fire took place; however, its topography and vegetation were like those of the reference watershed. Samples from the Rocky Fire were collected from Cache Creek at a location (38°55′24.83″N, 122°19′35.32″W) that captured the burned watershed area below Clear Lake. In general, the water in Cache Creek mainly originated from the watershed area below Clear Lake (shown in Fig. 1), as the water was impounded in the winter for subsequent release for spring-summer irrigation. Grab samples were collected from a well-mixed stream section for selected storm events and base flow conditions. Representative flow data were available from USGS gauging station 11451800, located downstream of the water quality sampling point (Fig. S2b). The difference in flow between the Clear Lake outflow (USGS station 11451800) and the downstream flow station was used to calculate flow generated from the watershed area containing the wildfire. Flow data indicated that the water sampling point mainly received runoff from the burned watershed area (the ratio of the lake discharge/sampling point flow was <0.5%) during initial flushes of the first rainy season (Fig. S2c). Therefore, the proportion of burned areas below Clear Lake contributing to the sampling point was ~40% during the initial flushes. However, the averaged contribution of water released from the lake was 26% and 48% during the sampling periods corresponding to the subsequent flushes and the second rainy season’s flushes, respectively. Consequently, the impact of the Rocky Fire was attenuated by dilution, especially during the later portion of each annual water collection period.

2.3. Analytical methods

2.3.1. Water quality analysis and DOM characterization

Water samples were collected in pre-rinsed/baked amber glass bottles (1000 mL) and delivered chilled (ice packs) to the Clemson water quality laboratory within 72 h. Separate subsamples were filtered through pre-combusted glass fiber (Whatman 934-AH, ~0.7 μm pore size) and membrane filters (Supor®, PES 0.45 μm pore size) and kept at 4 °C through the completion of analysis. Determination of DOC, total dissolved nitrogen (TDN), ammonia (NH_4^+ —N), nitrate (NO_3^- —N), nitrite (NO_2^- —N), and bromide (Br^-) and $SUVA_{254}$ (describing the aromaticity of DOM) is explained elsewhere (Majidzadeh et al., 2020; Ruecker et al., 2017), in the Supporting Information (SI), and in Table S1. Dissolved organic nitrogen [DON] was calculated as the difference between TDN and inorganic-N ($NH_4^+ + NO_3^- + NO_2^-$). Fluorescence excitation-emission matrices (EEMs) and the associated parameters including fluorescence regional integration (I: tyrosine-like, II: tryptophan-like, III: fulvic acid-like, IV: soluble microbial byproduct-like and V: humic acid-like), humification index (HIX), fluorescence index (FI), and freshness index (β:α) were calculated as previously described (Chen et al., 2003; Ruecker et al., 2017; Zhou et al., 2013).

2.3.2. DBP formation potential (FP) tests

DBP-FP tests were conducted to determine maximum precursor concentration levels in chlorinated and chloraminated water samples. For four THMs (trichloro-, dichlorobromo-, dibromochloro- and tribromomethane), nine HAAs (chloro-, bromo-, dichloro-, bromochloro-, trichloro-, dibromo-, bromodichloro-, dibromochloro-, and tribromo acetic acid) and chloral hydrate (CH) FPs, water pH was adjusted to 7.8 ± 0.1 with 10 mM borate buffer and then oxidized with excess Cl_2 (as NaOCl) as determined from the formula: $Cl_2 = 3x[DOC]+7.6x[NH_4^+]+10$, where [DOC] and $[NH_4^+]$ were in mg/L (Krasner et al., 2004). For NDMA FP, glass-filtered water samples were adjusted to 7.8 ± 0.1 with 10 mM borate buffer and oxidized with 100 mg/L pre-formed monochloramine (NH_2Cl) (Cl_2/N ratio of 4:1) (Jones et al., 2012). Excess oxidant-added, space-free samples were kept in pre-washed amber bottles for five days (at room temperature [21 ± 2 °C]) in the dark. After incubation, residual oxidant was measured in all samples (oxidant content was always >10 mg/L and >20 mg/L for Cl_2 and NH_2Cl, respectively) and quenched with a stoichiometric amount of ascorbic acid (for chlorination) or sodium thiosulfate (for chloramination) before extraction for DBP determination. Detailed descriptions for analysis of general water quality parameters, DOM characterization, analysis of DBPs, and minimum reporting levels (MRLs) are presented elsewhere (Uzun et al., 2017; Ruecker et al., 2017), and summarized in the SI. Student's t-tests were used to determine significant differences among watersheds. Box-and-whisker plots were used to depict selected data graphically and provide the maximum, minimum, 90th, 75th, median, 25th, and 10th quartile metrics (Fig. S3).

3. Results and discussion

3.1. Release of suspended solids

Post-fire rainstorms resulted in the release of high amounts of eroded particles from barren soils of the burned areas. During the first rainy season in the unburned reference watershed, measured values (average ± std.) were 36 ± 47 NTU, 362 ± 478 Pt—Co, and 96 ± 158 mg/L for turbidity, apparent color, and total suspended solids (TSS), respectively (Figs. S4a, b, and c). In contrast, burned areas saw significantly higher ($p < 0.05$) values (871 ± 747 and 4478 ± 6720 NTU, 6014 ± 6098 and 28316 ± 40248 Pt—Co, and 613 ± 626 and 4795 ± 6237 mg/L for the Rocky and Wragg Fires, respectively) during the first rainy season initial flushes, but decreasing trends (1337 ± 1775 and 137 ± 138 NTU, 6522 ± 8266 and 3399 ± 4272 Pt—Co, and 1066 ± 1303 and 700 ± 1127 mg/L for the Rocky and Wragg Fires, respectively) during the first rainy season's subsequent flushes, especially for the Wragg Fire. TSS vs. turbidity and apparent color showed strong positive linear correlations ($R^2 \geq 0.9$) for all watersheds (Table S2). During the second rainy season's flushes, measured values for all three constituents increased ($p < 0.05$) in the reference site (Figs. S4a, b, and c). Higher precipitation (431 vs. 1029 mm) in the second rainy season likely caused higher erosion and particle transport in the reference watershed. For the Rocky Fire, values (turbidity, color, and TSS) were comparable between the first and second years. As only a portion of the Cache Creek watershed was burned by the Rocky Fire, the water quality response was not as strong as that found in the Wragg watershed, which was more extensively burned (>90%). Second-year values for the Wragg Fire showed a remarkable decrease with values like the reference site. The sharp decrease in suspended sediments resulted from rapid erosion of the ash layer in the first year followed by decreased erosion due to rapid revegetation of the burned watershed by stump-sprouting shrubs and annual grasses in the second year. These findings are consistent with other studies that show distinct flushing of sediments, color, and TSS during the initial storm events following wildfires (Emelko et al., 2011; Hohner et al., 2018, 2016; Mast et al., 2016; Murphy et al., 2015, 2012; Son et al., 2015; Writer et al., 2014).

Quick mobilization of fire-generated particles into the watershed increased concentrations of suspended solid related parameters, particularly during the initial storm events. On the other hand, the first post-fire rainy season had total dissolved solids (TDS) (calculated by subtracting TSS concentrations from total solids [TS]) averaged values: 96 ± 156, 262 ± 109, and 329 ± 456 mg/L in the reference, Rocky and Wragg Fire samples, respectively (Fig. S4d). However, all averaged values were lower than the EPA secondary drinking water standard of 500 mg/L (USEPA, 2012). The TDS/TS ratios in the Rocky (24%) and Wragg Fire (10%) samples were significantly lower ($p < 0.05$) than those in the samples from the reference watershed (73%) (Table S3) indicating higher erosion rates in burned watersheds for the first post-fire rainy season. During the second post-fire rainy season, the averaged TDS/TS decreased in the reference watershed (from 73% to 24%) ($p < 0.05$). However, the TDS/TS ratios in reference (24%) samples were lower compared to those in the Rocky (43%) and Wragg Fire (37%) samples. These data indicate increased erosion in the reference watershed, and quick mobilization of fire-generated suspended solids during the first rainy season, especially in the more extensively (>90%) burned watershed.

3.2. Release of aromatic carbon

During the first post-fire rainy season, DOC concentrations were not statistically different ($p > 0.05$) in reference (5.4 ± 2.6 mg/L) vs. Rocky Fire samples (4.9 ± 1.6 mg/L) (Fig. 2a). However, the Wragg Fire samples had significantly higher ($p < 0.05$) DOC concentration (9.0 ± 2.2 mg/L) during the first rainy season's initial flushes compared to the reference and Rocky Fire samples. It has been indicated that the production of mg-Carbon/g-detritus materials is highly dependent on fire severity (Wang et al., 2015). Therefore, we may expect a significant decrease in the release of DOC after high—severity fires. However, the release of DOC from Wragg Fire was higher than that released from the reference during the initial flushes. This is consistent with the predominantly medium severity of Wragg Fire that resulted in higher DOC release and mobility from the more mobile, ash derived carbon pools generated from the

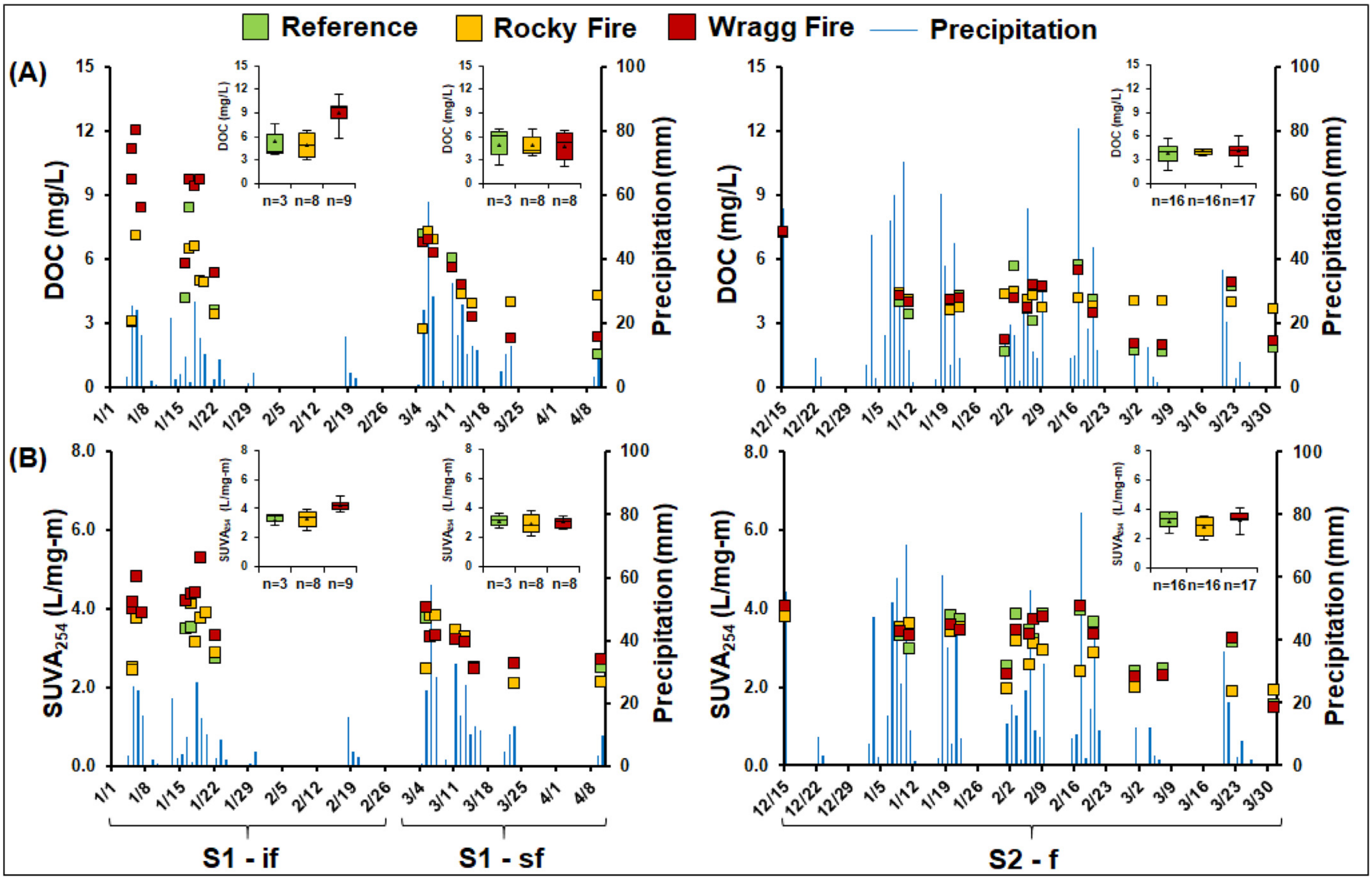

Fig. 2. Water quality characteristics in the reference, Rocky, and Wragg Fire samples for S1−if, S1−sf, and S2−f. DOC (A) and $SUVA_{254}$ (B). n: number of samples. S1-if and S1−sf define samples collected during the initial (Jan. 5−22, 2016) and subsequent flushes (Mar. 5 − Apr. 10, 2016) for the first rainy season, respectively. S2−f defines all samples collected during the second rainy season (Dec. 15, 2016−Mar. 31, 2017). Streamflow generation was delayed in the Reference watershed each year compared to the burned watersheds.

burned watersheds during the initial flushes. During subsequent rain events, the DOC concentration in the reference and Rocky Fire samples did not change significantly ($p > 0.05$) (4.9 ± 3.0 mg/L vs. 4.9 ± 1.6 mg/L). However, significant decreases ($p < 0.05$) were observed (from an average of 9.0 to 4.9 mg/L) compared to those of initial flushes in the Wragg Fire samples. This observation indicates that fires can increase the release of DOC, but the response will be more apparent as the percentage of the burned watershed area increases. During the second rainy season, the averaged DOC values decreased to 3.8−4.2 mg/L across all watersheds (Fig. 2a). This observation is consistent with previous findings where recently generated ash had higher DOC leaching potential than did weathered/leached ash (Wang et al., 2016). Overall, these findings indicate that while fires consume large amounts of litter/vegetation, they can generate more mobile DOC that is quickly flushed (<1 year) from the burned watersheds in the post-fire runoff.

For the first rainy season, in general, the average $SUVA_{254}$ did not change significantly ($p > 0.05$) and overlapped for all sample groups (Fig. 2b). During the initial flushes, the $SUVA_{254}$ values were 3.2 ± 0.4 and 3.3 ± 0.7 L/mg-m for the reference and the Rocky Fire samples, respectively. However, the Wragg Fire samples only showed 31 ± 17% higher (4.5 ± 0.6 L/mg-m) values that decreased to 3.1 ± 0.5 L/mg-m (similar to reference and Rocky Fire) over subsequent rain events. Higher $SUVA_{254}$ values suggest greater aromaticity of the DOM fraction (Weishaar et al., 2003) during the initial flushes. Previous research found that ash materials (Abraham et al., 2017) and polycyclic aromatic hydrocarbon (PAH) structures form during wildfires (Vila-Escalé et al., 2007), and these newly generated compounds can increase the hydrophobicity of water and soil materials. Moreover, Wang et al. (2015) showed that the $SUVA_{254}$ values of the water leachate of ash materials follow the order: white ash (high temperature) > unburned detritus > black ash (cooler temperature). Thus, the higher ($p < 0.05$) $SUVA_{254}$ in the Wragg Fire samples during the first rainy season's initial flushes may be related to the newly generated white ash and PAHs that reach peak concentration in recently burned sites (Tsibart et al., 2014), and decrease over time with precipitation and leaching (Olivella et al., 2006; Vila-Escalé et al., 2007). Thus, the decreasing trend in aromaticity of the Wragg Fire samples during the first rainy season's subsequent flushes may be related to the rapid removal by erosion/leaching of these newly generated aromatic/hydrophobic compounds. By the second rainy season, the average $SUVA_{254}$ values for the reference and burned watersheds were comparable (3.2 ± 0.7, 2.8 ± 0.7 and 3.3 ± 0.8 L/mg-m for reference, Rocky and Wragg Fire samples, respectively) (Fig. 2b). This confirms that the fire-generated aromatic compounds flushed quickly from burned watersheds and there were no significant seasonal changes in leaching of those newly generated materials.

Soil pH increases after wildfire due to the mineralization and release of acidic volatiles (Certini, 2005; Wang et al., 2015, 2016), and the concentration of Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+} as oxides, hydroxides, and carbonates (Certini, 2005), especially in newly burned organic matter (Revchuk and Suffet, 2014). Therefore, post-fire stream runoff may increase the pH of downstream surface waters (Son et al., 2015), possibly resulting in a higher solubility of DOM. However, the length of pH enhancement was variable (up to

3 years) (Certini, 2005; Granged et al., 2011; Oliver et al., 2012; Wang et al., 2015). In our study, average stream water pH values were not significantly different ($p > 0.05$) for burned vs. reference samples (pH range = 7.91–8.33) (Fig. S5). Findings from our study indicate that several factors (e.g., burning severity, mobility of newly generated DOM, soil buffering capacity, weathering conditions) play a role in regulating the pH of stream water in burned watersheds.

3.3. Fire affects the nitrogen cycle

Nitrogen plays a vital role for all watershed organisms (Wilhelm, 2009), but increased nutrients (i.e., N, P) in downstream water bodies can cause adverse effects, such as eutrophication and harmful algal blooms (Bladon et al., 2014; Emelko et al., 2016; Santín et al., 2018). During the first post-fire rainy season, TDN consisted mainly (98%, 92% and 90% in the reference, Rocky and Wragg Fire samples, respectively) of organic nitrogen. In the reference samples, average TDN (data not shown), DON, and NH_4^+-N concentrations were 0.38, 0.33, 0.05 mg/L, respectively. However, those values were significantly higher ($p < 0.05$) in the fire impacted samples (Fig. 3a and b). In contrast, the NO_2^- (data not shown) and NO_3^- (Fig. 3c) concentrations were generally lower than the MRLs (presented in SI) in all watersheds. Notably, the DON concentrations in the Wragg Fire samples were significantly higher than those in the Rocky Fire samples. This likely reflects the higher percentage of burned areas occurring in the Wragg Fire watershed. Despite lower TDN and DON leaching capacities from ash (Wang et al., 2016) and soil samples (Homann et al., 2011) found in some studies, elevated concentrations of TDN, DON and NH_4^+ have been reported by others in stream waters following wildfires (Bladon et al., 2008; Hohner et al., 2016; Murphy et al., 2015; Son et al., 2015). These nitrogen forms tend to accumulate in the ashy materials following release by pyromineralization of vegetation/litter and are quickly mobilized and transported via surface runoff/erosion following wildfires (Knicker, 2010; Knicker et al., 2005; Mast et al., 2016; Neary et al., 2005). Additionally, the DOC/DON ratio was significantly lower ($p < 0.05$) in the Rocky (5.0 ± 1.63) and Wragg Fires (4.9 ± 1.85) samples compared to reference (17.4 ± 5.87) (Fig. S6). This lower DOC/DON ratio from burned watershed samples is consistent with a laboratory study where a lower DOC/DON ratio resulted from wildfire ash leachates compared to a control (Hohner et al., 2016; Ranalli, 2004; Wang et al., 2015).

In the second-year post-fire, both the TDN (data not shown) and DON concentrations decreased in all samples compared to the first rainy season, most likely as a result of dilution due to much higher precipitation in year two. However, both the TDN and DON concentrations remained higher in the burned watersheds relative to the reference watershed. This indicates that TDN and DON continued to leach from the burned sites for extended periods (>1 year). The average NO_3^- concentrations were highest for the second rainy season (Fig. 3c), consistent with several previous studies (Koyama et al., 2010; Mast et al., 2016; Murphy et al., 2015; Smith et al., 2011). A lower DOC/DON ratio (Fig. S6) and mineralization of organic matter from the ash/soil leads to the accumulation of NH_4^+ as plant uptake is attenuated following the fire. These conditions provide a favorable environment for nitrifying microorganisms to thrive and generate NO_3^-, which is highly mobile and susceptible to leaching. Given the slow growth of nitrifying microorganisms under the cold winter conditions, NO_3^- generation and leaching were hindered in the burned areas in the first year following wildfire (Acea and Carballas, 1996). In summary, in the first year following wildfire, there was a decrease in the DOC/DON ratio and an increase in the release in NH_4^+ and DON, which made up the majority of TDN. These conditions probably stimulated nitrification, which in turn resulted in delayed transport of NO_3^- from the watershed in the second year following the wildfire. This was consistent with a recent study where the nitrate level was an order of magnitude higher in burned catchment stream-water compared to unburned catchment stream-water (Rhoades et al., 2019).

3.4. Changes in DOM composition following wildfire

EEM regions and fluorescence indices provide information about DOM sources, chemical characteristics, and dynamics in aquatic ecosystems (Fellman et al., 2010), and wildfire can alter the EEM fluoresce regions of DOM (Revchuk and Suffet, 2014; Wang et al., 2015). Considering the potential interference of metals, (e.g., iron) on optical measurements, such as $SUVA_{254}$ and fluorescence measurements (Poulin et al., 2014; Weishaar et al., 2003), we analyzed only samples having iron concentrations <0.03 mg/L. Thus, the number of measurements ($n = 2$) for the reference watershed for the first rainy season was limited. As a result, it was not possible to apply statistical analysis between reference and burned watershed samples, which limits a definitive conclusion. Although the DOC content and sample collection periods (4.1 and 8.4 mg/L for initial flushes and subsequent flushes, respectively) were different for those two samples, DOM characteristics were similar (Table 1). Considering also a minimal variability of the second-year measurements ($n = 6$), we concluded that the DOM character did not significantly change in the reference samples. Therefore, we interpreted our results accordingly. Results showed that EEM region contributions for the reference samples were dominated by humic acid-like DOM (56%) during the first post-fire rainy season. In contrast, the Rocky and Wragg Fire samples were distinctly different from reference samples for the first runoff event (for the Rocky Fire, $n = 3$; for the Wragg Fire, $n = 4$). During the first rainy season's initial flushes, humic-like and fulvic-like components contributed 31–33% of DOM, while tryptophan-like and tyrosine-like components contributed 16% and, 2–4% of DOM. In the burned watershed samples, after 3–4 flushes for the percentage of humic-like DOM came to a level comparable to the level in the reference; this process was rapid compared (2 years) to a previous study (Revchuk and Suffet, 2014). Also, DOM from burned watersheds had significantly higher ($p < 0.05$) fulvic acid- and protein-like components, as well as lower soluble microbial by-product-like (SMP) compositions. This suggests the formation/release of fulvic-like DOM and protein-like DOM (Fellman et al., 2010), as well as, lower molecular weight (MW) microbial DOM sources resulting from the fire and/or post-fire biological activity (Revchuk and Suffet, 2014). Higher tyrosine-like and especially tryptophan-like regions indicated that amino acids and aromatic proteins, mainly intact proteins and less degraded peptides, were generated by post-fire biological activity (Chen et al., 2003; Fellman et al., 2010). However, these compounds were likely flushed from the watershed during the initial 3–4 flushes, contributing to the higher DON exported from the burned watersheds (Fig. 3A).

Averaged fluorescence index (FI) values for the reference and Rocky Fire samples were similar (1.55 ± 0.01 [$n = 2$] and 1.57 ± 0.03 [$n = 7$], respectively); however, the Wragg Fire samples had higher values (1.70 ± 0.04 [$n = 8$]) during the first flush (Table 1). Unlike previous work (Hohner et al., 2016), our results suggest extensive wildfires can increase FI value. The humification index (HIX) and Freshness index (β/α) for the reference samples were 8.43 ± 1.90 and 0.65 ± 0.01 ($n = 2$), respectively. In contrast, the Rocky and Wragg Fire samples had higher HIX and β/α values during the initial flushes (for Rocky, 10.22 ± 3.02 and 0.81 ± 0.02 [$n = 3$]; for the Wragg, 24.35 ± 7.50 and 0.89 ± 0.01 [$n = 4$]). These elevated values

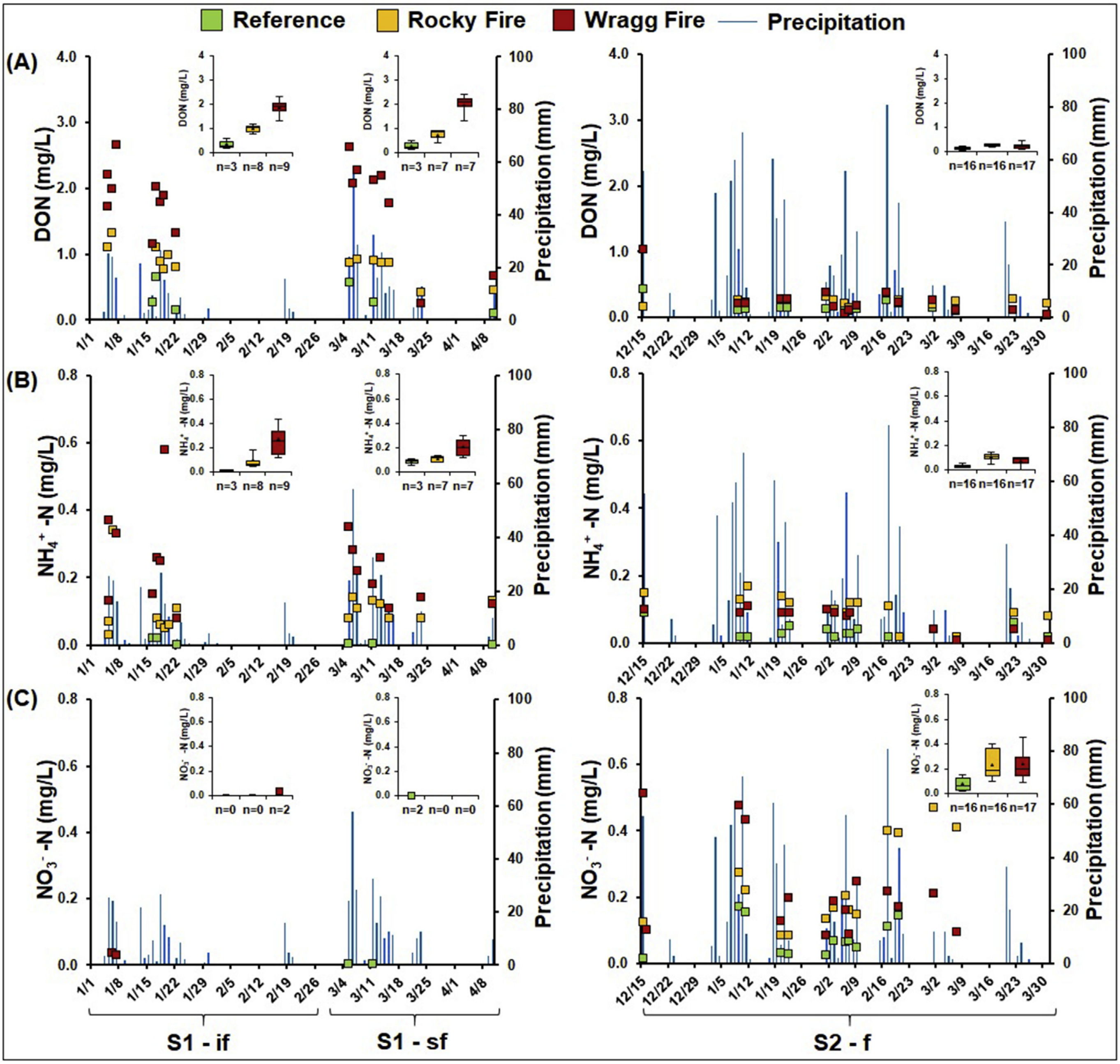

Fig. 3. Changes in nitrogen forms in the reference, Rocky Fire, and Wragg Fire samples for S1—if, S1—sf, and S2—f. DON (A), Nitrate-N (B), and Ammonia-N (C). n: number of samples. S1-if and S1—sf defines samples collected during the initial (Jan. 5—22, 2016) and subsequent flushes (Mar. 5 — Apr. 10, 2016) for the first rainy season, respectively. S2—f defines all samples collected during the second rainy season (Dec. 15, 2016—Mar. 31, 2017). Streamflow generation was delayed in the Reference watershed each year compared to the burned watersheds.

decreased slowly with subsequent rainfall/runoff events. Considering the change in the DOM composition, these observations indicate that wildfires can consume much of the terrestrial DOM sources, and enhanced microbial activity from residuals may generate lower MW of DOM in the burned watersheds (Revchuk and Suffet, 2014; Tsai et al., 2017; Valenca et al., 2020; Wang et al., 2015). In addition, the newly formed DOM is likely more mobile, resulting in its rapid flushing from the watershed.

For the second rainy season, the EEM average humic-like region (78%) increased significantly across all watersheds, and FI, HIX and β/α values were similar across burned and reference watersheds. Thus, the DOM spectroscopic properties indicate that wildfire has a pronounced short-term (≤1 year) effect on DOM quality, but DOM quickly reverts to characteristics like the reference watershed in the second-year post-fire.

3.5. DOC governs THM and HAA precursor concentrations

During the first post-fire rainy season, average THM, HAA, and CH FP concentrations in the reference and the Rocky Fire samples were 540 ± 339, 355 ± 208, and 73 ± 52 μg/L and 504 ± 236, 360 ± 223, and 45 ± 34 μg/L, respectively. Notably, the Wragg Fire samples had higher average THM and HAA concentrations of 687 ± 279 and 616 ± 421 μg/L, respectively (Fig. 4a, b, and c), but

Table 1
Selected DOM characteristics for the first and the second rainy seasons.

			DOC (mg/L)	Tyrosine-like (%)	Tryptophan-like (%)	Fulvic acid-like (%)	SMP-like (%)	Humic acid-like (%)	FI	HIX	β/α
The first post-fire rainy season	Reference	All (n = 2)	6.3 ± 2.2	1.7 ± 0.6	5.5 ± 0.5	13.8 ± 0.1	23.2 ± 1.3	55.8 ± 2.3	1.55 ± 0.01	8.4 ± 1.9	0.65 ± 0.00
	Rocky Fire	Initial flushes (n = 3)	4.4 ± 2.3	4.1 ± 1.3	16.0 ± 1.6	29.8 ± 1.0	18.5 ± 0.9	31.6 ± 2.6	1.57 ± 0.02	10.2 ± 3.0	0.81 ± 0.02
		Subsequent flushes (n = 4)	5.7 ± 0.9	1.1 ± 0.2	5.8 ± 0.5	13.5 ± 0.5	25.7 ± 2.9	54.0 ± 2.8	1.58 ± 0.04	8.8 ± 1.7	0.76 ± 0.06
	Wragg Fire	Initial flushes (n = 4)	10.3 ± 1.6	2.1 ± 0.4	15.6 ± 0.4	29.9 ± 0.4	19.7 ± 0.3	32.8 ± 0.2	1.71 ± 0.03	24.4 ± 7.5	0.89 ± 0.01
		Subsequent flushes (n = 4)	8.6 ± 1.9	0.4 ± 0.3	5.8 ± 1.0	12.7 ± 0.3	28.3 ± 2.4	53.0 ± 3.3	1.69 ± 0.06	10.5 ± 4.9	0.86 ± 0.05
Second post-fire rainy season	Reference	All (n = 6)	3.7 ± 1.4	0.5 ± 0.1	1.6 ± 0.3	10.3 ± 0.3	9.2 ± 2.0	78.4 ± 2.0	1.50 ± 0.03	6.4 ± 1.0	0.70 ± 0.09
	Rocky Fire	All (n = 7)	4.0 ± 0.3	0.6 ± 0.1	1.7 ± 0.2	10.3 ± 0.2	9.5 ± 1.4	77.9 ± 1.6	1.52 ± 0.04	6.4 ± 2.0	0.74 ± 0.06
	Wragg Fire	All (n = 6)	4.3 ± 0.4	0.5 ± 0.2	1.6 ± 0.3	10.3 ± 0.4	9.0 ± 2.2	78.6 ± 2.4	1.51 ± 0.06	8.2 ± 2.0	1.71 ± 0.06

Mean ± standard deviation, n: number of samples. Values for reference samples (the first post-fire rainy season only) were calculated taking the difference between two measurements. Iron content in the samples was <0.03 mg/L in all samples.

similar values for CH FPs (68 ± 43 μg/L) compared to both the reference and Rocky Fire samples. Correlations for DOC vs. THM and HAA FPs were strongly positive ($R^2 \geq 0.7$), while DOC vs. CH FP correlations was moderately positive ($0.5 \leq R^2 \leq 0.7$) (Table S4) (Rumsey, 2016). Therefore, the higher THM and HAA FPs were associated with higher DOC concentrations (~40%) in the Wragg Fire samples (Fig. 2a). DOC normalized concentrations were further calculated to examine the reactivity of DBP precursors per unit of carbon. Results showed that the carbon normalized average reactivity and variability of THM and HAA precursors were within a similar range for all samples (THM FP/DOC: 99 ± 18 μg/mg-C, HAA FP/DOC: 70 ± 27 μg/mg-C). These data are consistent with previous studies that showed that the carbon-normalized reactivity of C-DBP precursors was not significantly affected by wildfire (Hohner et al., 2016; Writer et al., 2014). Therefore, the increase in C-DBP precursor concentrations (Cawley et al., 2016; Revchuk and Suffet, 2014) was due mainly to the enhanced mobility of DOC, especially during the first post-fire runoff events. In contrast, carbon-normalized CH FP values decreased (~30%) in the Rocky and Wragg Fire samples compared to the reference samples. Previously, Wang et al. (2016) showed that DOC and CH FP yields (μg/g-detritus) for wildfire ash samples were lower compared to their unburned source materials. Thus, alteration of DOC quantity and quality both appear to affect the reactivity of CH precursors.

Results further showed that NDMA FP values (27 ± 8.6 and 28 ± 16.0 ng/L for Rocky and Wragg Fire samples, respectively)

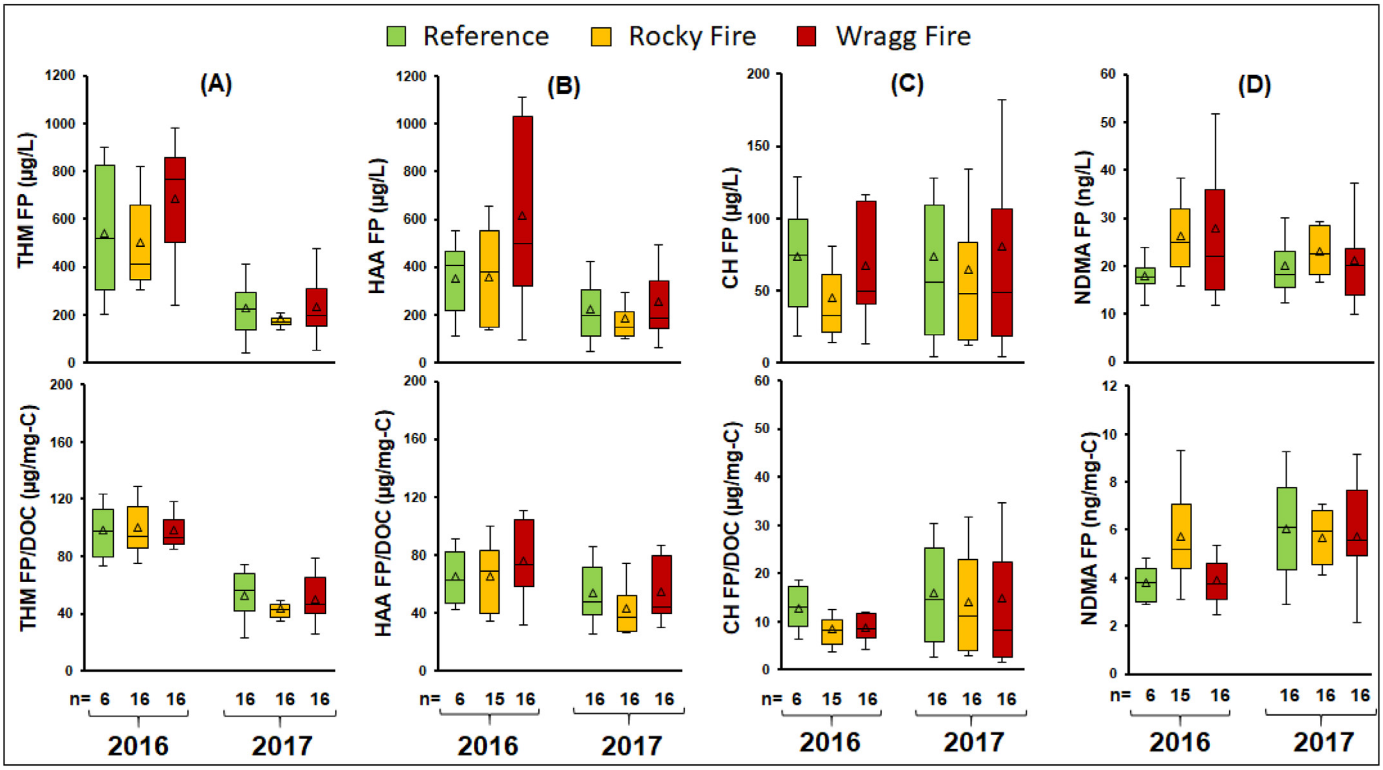

Fig. 4. FP and carbon normalized DBP FP values in the reference, Rocky Fire, and Wragg Fire samples for samples collected in 2016 and 2017, respectively. THM FPs (A), HAA FPs (B), CH FPs (C), and NDMA FPs (D). n: number of samples.

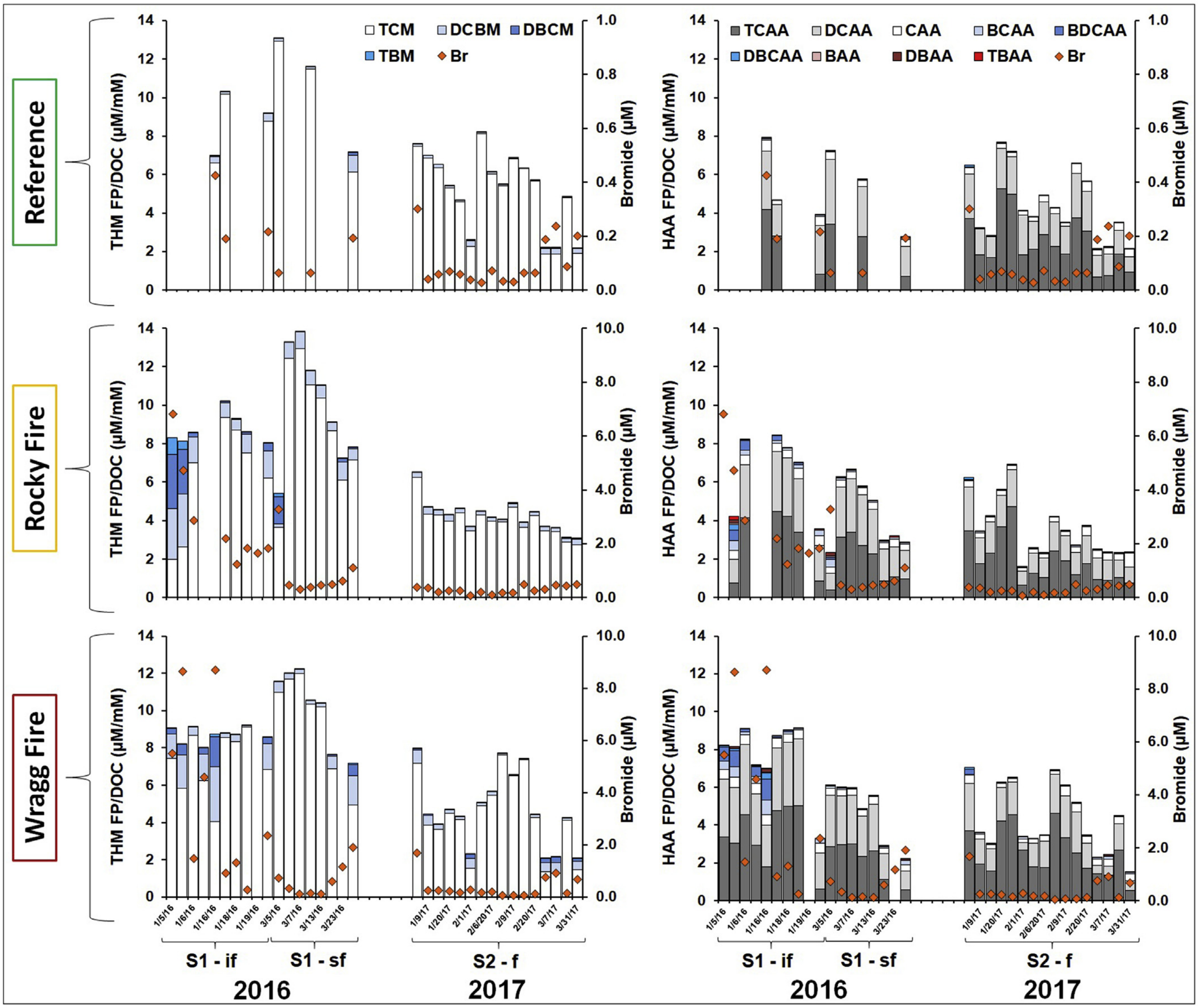

Fig. 5. Speciation for carbon-normalized DBP FPs (THM/DOC and HAA FP/DOC) and bromide ion concentrations for S1−if, S1−sf, and S2−f. S1-if and S1−sf define samples collected during the initial (Jan. 5–22, 2016) and subsequent flushes (Mar. 5–Apr. 10, 2016) for the first rainy season, respectively. S2−f defines all samples collected during the second rainy season (Dec. 15, 2016–Mar. 31, 2017).

were significantly ($p < 0.05$) higher in the burned watershed samples compared to reference samples (18 ± 5.9 ng/L) (Fig. 4d). These results are consistent with studies by Wang et al. (2015, 2016) that showed ash sample leachates had relatively higher NDMA FP compared to their unburned source materials. Since NDMA FP did not correlate with DON or TDN (Uzun et al., 2015) and several known precursors are anthropogenic-derived (Ruecker et al., 2017; Uzun et al., 2017, 2015), such an increase may be explained by the use of human-made compounds, such as fire suppressants during the fire or the release of some of the NDMA precursors by high-temperature burns. Also, carbon-normalized NDMA FP values were higher ($p < 0.05$) in the Rocky Fire samples (5.8 ± 2.4 ng/mg-C) compared to Wragg Fire samples (3.9 ± 1.07 ng/mg-C) during the first rainy season. This may be related to more anthropogenic activity in the Cache Creek watershed (Rocky Fire). In addition, correlations between NDMA FPs and nitrogen species (DON and inorganic nitrogen) were weak ($R^2 \leq 0.5$).

During the second post-fire rainy season, significant decreases were observed in THM and HAA FPs in all watersheds, as well as carbon normalized reactivity. Values were similar across both burned and unburned samples, indicating that the wildfire effects were short-term. CH FP levels were similar to those in samples from the previous rainy season. Considering DOC trends and linear correlations between DOCs and C-DBPs (Table S4), the lower THM and HAA FPs could be due to lower DOC concentrations resulting from the greater rainfall amounts/dilution in the second year. However, the decrease in normalized carbon values cannot be explained by dilution, suggesting that some changes in DOM characteristics (ratio changes especially for fulvic acid-like vs. humic acid-like DOM) which caused lower DBP yields as well. The NDMA FP levels were similar between years for all watersheds, but the normalized carbon values were relatively higher in the second year. Considering the lower DOC levels in the second year, this finding further confirms that DOC is not a good proxy for NDMA precursors (Bond et al., 2011; Uzun et al., 2015; Yang et al., 2015).

3.6. *Formation of more toxic brominated THMs and HAAs*

In this section, bromine incorporation factors (BIFs) are calculated (Krasner et al., 2008) and discussed for THMs and HAAs. We present the concentrations for the bromide, DOC, THM, and HAA FPs in molar units. Bromide concentrations in the reference samples were significantly lower (0.16 ± 0.15 μM; n = 6) (Fig. 3d) than those found in the Rocky Fire (1.35 ± 0.9 μM; 2.96 ± 1.81 [n = 9, initial nine flushes] and 0.54 ± 0.26 [n = 7, subsequent seven flushes]) and Wragg Fire samples (1.67 ± 2.29 μM; 4.05 ± 3.56 [n = 9, initial nine flushes] and 0.65 ± 0.64 μM [n = 8, subsequent eight flushes]) (Fig. 5). Wang et al. (2015) showed that increased fire severity enhanced the yield of Br$^-$ from ash leachates as a higher degree of pyromineralization concentrates Br salts in the ash materials. However, other studies reported low (<0.125 μM) Br$^-$ concentration in samples collected from burned regions (Hohner et al., 2018, 2016). The main differences between these studies may be related to geology and proximity to the ocean, the primary source affecting precipitation chemistry. Our study site in northern California is located near the ocean, and the geologic materials are derived from marine sedimentary rocks, presumably containing relatively high Br concentrations. During the second rainy season, Br$^-$ concentrations were <MRL (0.125 μM), 0.26 ± 0.13, and 0.40 + 0.46 μM in the reference, Rocky Fire, and Wragg Fire samples, respectively (Fig. 5). The decreased Br$^-$ concentrations reflect dilution from the higher precipitation amounts in the second season, as well as leaching of Br$^-$ ion from the ash layer. Therefore, we concluded that the Br$^-$ release from burned watersheds could be high during initial post-fire runoff events and could slowly decline to background levels with subsequent runoff/leaching events.

During the first post-fire rainy season, trichloromethane (TCM) and chlorinated HAA species (chloro- [C], trichloro- [TC] and dichloro- [DC] acetic acids [AA]) accounted for >95% of total THM and HAA FPs on a molar basis in the reference site (Fig. 5). However, in parallel with Br$^-$ trends, THMs and HAAs showed changes in their distribution of chlorinated and brominated species in burned watersheds. In the Rocky Fire samples, for initial (Jan. 5–22, 2016) vs. subsequent flushes (Mar. 5–Apr. 10, 2016), TCM and chlorinated HAAs species consisted of 72 vs. 89% and 89 vs. 94% of total THM and HAA FPs, respectively. Similarly, in the Wragg Fire samples, TCM and chlorinated HAA species consisted of 83% vs. 89% and 88% vs. 94% of total THM and HAA FPs. These results indicate that brominated species were more predominant in the first rainy season's initial flushes having higher Br$^-$ concentrations (Fig. 5). BIF increased due to higher Br$^-$ content in the Rocky and Wragg Fire samples for both THM and HAA FPs (Figs. S7a and b). Similar to Br$^-$ trends for the second rainy season's flushes, BIF values in the burned watersheds remained higher compared to those in the reference site, but overall the concentration of brominated species was diminished as Br$^-$ concentrations decreased. These results show that BIF could increase following wildfires (Wang et al., 2015), and the effect can persist for multiple years (Chow et al., 2019). However, this observation requires further (using leachate water of unburned vs. burned materials with low vs. high bromide content) laboratory tests.

4. Conclusion

Wildfires can cause rapid and severe impairments in water quality during post-fire runoff events. Our study showed that the proportion of the watershed burned significantly increase the detrimental effects of downstream water quality. In the short term (<1 year), significantly higher turbidity, color, and suspended particles might be expected, and the degree of impairment is proportional to the amount of watershed area burned. Further, increased concentrations of DOC (in the short term [<1 year]), TDN, DON, and NH_4^+ are expected to be prevalent in the first-year runoff, along with changes in DOM chemical properties over the short to medium-term (up to 2 years). NO_3^- concentration could be negligible in the first-year runoff but appreciably higher concentrations are expected during the following year's post-fire runoff events, possibly due to delayed nitrification.

C-DBP precursor concentrations can be considerably higher due to the release of more mobile and more aromatic DOC from the catchment in the short term (<1 year), but the C-normalized reactivity of DBP precursors is not expected to be altered significantly by wildfire. Elevated Br$^-$ concentrations can be found especially in burned watersheds located in coastal regions. Thus, more effective treatment processes/applications may be required to limit the formation of more toxic brominated DBP in fire impacted treated drinking water. Therefore, chloramination can be considered as a secondary disinfection process to treat fire-impacted raw water, if the water utility is using high DOC content source waters, and its typical THM and HAA levels are close to the limits in their distribution systems. Finally, utilities must be prepared for elevated levels of suspended solids, relatively higher DOC, unusual nitrogen (different speciation), and relatively higher bromide content in their raw waters.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2020.115891.

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