

Sediment-phosphorus dynamics can shift aquatic ecology and cause downstream legacy effects after wildfire in large river systems

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Abstract

Global increases in the occurrence of large, severe wildfires in forested watersheds threaten drinking water supplies and aquatic ecology. Wildfire effects on water quality, particularly nutrient levels and forms, can be significant. The longevity and downstream propagation of these effects as well as the geochemical mechanisms regulating them remain largely undocumented at larger river basin scales. Here, phosphorus (P) speciation and sorption behavior of suspended sediment were examined in two river basins impacted by a severe wildfire in southern Alberta, Canada. Fine-grained suspended sediments (<125 μm) were sampled continuously during ice-free conditions over a two-year period (2009–2010), 6 and 7 years after the wildfire. Suspended sediment samples were collected from upstream reference (unburned) river reaches, multiple tributaries within the burned areas, and from reaches downstream of the burned areas, in the Crowsnest and Castle River basins. Total particulate phosphorus (TPP) and particulate phosphorus forms (nonapatite inorganic P, apatite P, organic P), and the equilibrium phosphorus concentration (EPC₀) of suspended sediment were assessed. Concentrations of TPP and the EPC₀ were significantly higher downstream of wildfire-impacted areas compared to reference (unburned) upstream river reaches. Sediments from the burned tributary inputs contained higher levels of bioavailable particulate P (NAIP) – these effects were also observed downstream at larger river basin scales. The release of bioavailable P from postfire, P-enriched fine sediment is a key mechanism causing these effects in gravel-bed rivers at larger basin scales. Wildfire-associated increases in NAIP and the EPC₀ persisted 6 and 7 years after wildfire. Accordingly, this work demonstrated that fine sediment in gravel-bed rivers is a significant, long-term source of in-stream bioavailable P that contributes to a legacy of wildfire impacts on downstream water quality, aquatic ecology, and drinking water treatability.

Keywords: climate change, cumulative watershed, effects, eutrophication, land disturbance, phosphorus, sediment, treatability, wildfire

Received 21 June 2015 and accepted 3 August 2015

Introduction

Wildfire is the dominant natural landscape disturbance in many forested landscapes, including much of western North America (Weber & Flannigan, 1997). Increased occurrence of large, severe wildfires and longer fire seasons have been attributed to climate change (Stocks *et al.*, 2002; Westerling *et al.*, 2006, 2011; Flannigan *et al.*, 2009). These increasing trends in wildfire are expected to continue due to increases in global mean temperature (IPCC, 2013; Flannigan *et al.*, 2013). Further increases in fire severity, the areal extent of burn, and wildfire season length in North American

forests are expected to continue through to the end of the century (Flannigan *et al.*, 2005). Similar predictions of more frequent, larger fires have been made for South America, central Asia, southern Europe, southern Africa, and Australia (Pechony & Shindell, 2010).

The effects of severe wildfire on terrestrial and aquatic ecosystems can be catastrophic, but the longevity of these effects varies among regions depending on the interaction of climate, physiography, geology, and vegetation (Shakesby & Doerr, 2006; Hauer *et al.*, 2007; Hurteau *et al.*, 2013; Moody *et al.*, 2013; Bladon *et al.*, 2014). It is well documented that sediment erosion rates increase after wildfire (Kunze & Stednick, 2006; Silins *et al.*, 2009a; Moody *et al.*, 2013). The delivery of sediment-associated phosphorus (P) from hill slopes to

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receiving streams can substantially increase P export to downstream environments; postfire P export has been reported at levels 0.3 to more than five times greater than at prefire conditions (Bayley *et al.*, 1992; Hauer & Spencer, 1998; Burke *et al.*, 2005; Lane *et al.*, 2008; Blake *et al.*, 2010; Silins *et al.*, 2014). While Lane *et al.* (2008) reported rapid postfire recovery in P export by the second year after wildfire in Australia, Spencer *et al.* (2003) reported that some effects on soluble reactive phosphorus (SRP) were still evident 3–4 years after wildfire in Montana. In contrast, Silins *et al.* (2014) observed no postfire recovery of sediment or P export, or associated effects on stream benthos (algae, stream invertebrates, and fish) a half decade after the Lost Creek wildfire in southwestern Alberta, Canada (also the study site herein). While downstream propagation of these effects might be expected depending on wildfire size and severity (Gresswell, 1999), these potential downstream cumulative watershed effects remain undocumented at larger river basin scales.

Sediment is the primary vector of P transport in aquatic systems (Kronvang *et al.*, 1997; House, 2003). Characterization of changes in aqueous P alone in aquatic ecosystems affected by land disturbance may not adequately describe disturbance impacts on P dynamics, because SRP released from sediment can be quickly utilized by primary producers. Stream sediments contain biogeochemical signatures that reflect the history of natural and human-impacted ecosystem processes in a watershed (e.g. erosion, mass wasting, weathering, etc.) (Meybeck, 1982). The physical and geochemical properties of bed and suspended sediment strongly influence the form (Stone & English, 1993) and mobility (Jarvie *et al.*, 2005) of particulate phosphorus (PP) in these systems. These materials contain different elements as a result of precipitation, exchange, and adsorption/desorption reactions that occur on, and within, the sediment structure (Hieltjes & Lijklema, 1980). The transport and bioavailability of P in aquatic environments is influenced by several physico-chemical factors that include the presence of organic compounds, particle size, temperature, pH, and redox conditions, as well as concentrations of major elements (e.g. Al, Fe, Mn, Ca, Mg) (Golterman, 2004). For example, the presence and oxidation state of Fe can drive solid-aqueous phase P equilibrium dynamics. P adsorption on iron hydroxide surfaces occurs more in aerobic environments due to complexation with Fe(III); however, P is released from sediments when solid Fe(III)-PO₄ forms are transformed due to the reduction of Fe(III) to Fe(II) (Golterman, 2004). Mn-Fe-polyphosphate and calcium phosphate complexes also commonly form (Golterman, 2004).

The various forms of particulate P are typically determined by sequential extraction techniques and defined operationally according to their ecological significance (Boström *et al.*, 1982; Pettersson *et al.*, 1988). Nonapatite inorganic phosphorus (NAIP) is comprised of phosphate adsorbed to noncalcium, metal hydroxide surfaces; at certain environmental conditions, it readily desorbs to the water column, making it the most bioavailable particulate P form (De Pinto *et al.*, 1981). Apatite phosphorus (AP) is considered geochemically stable and biologically unavailable (Pettersson *et al.*, 1988). Organic phosphorus (OP) is potentially bioavailable after hydrolysis and in some cases mineralization, but this refractory P is typically considered unavailable for primary productivity over relatively short temporal scales (Boström *et al.*, 1988a,b; Golterman, 2004). The relative abundance of particulate P forms can vary widely in river systems as a function of land use and disturbance. For example, sediments in rivers dominated by agriculture (Stone & English, 1993; Stone & Droppo, 1994; Fogal *et al.*, 1995) or urbanization (Owens & Walling, 2002; Ballantine *et al.*, 2008) typically have significantly higher levels of both NAIP and TPP compared to rivers that drain relatively undisturbed landscapes. Sharpley *et al.* (2014) described the cumulative effects of past land management practices on sediment-P dynamics in rivers as 'phosphorus legacy'. Although wildfire can be a significant, particularly transformative land disturbance, and despite the volume of literature regarding land use impacts on sediment-associated P dynamics in streams, wildfire impacts on particulate P forms and potential legacy effects in streams draining impacted landscapes, and higher order rivers downstream of wildfire-impacted areas, have not been previously reported.

Aquatic sediments play a critical role in buffering concentrations of SRP via sorption reactions between sediment and the water column (Froelich, 1988; Fox, 1993). Phosphate release from sediment into the water column depends upon several biophysical and chemical factors such as particle size, sediment geochemistry, settling velocity, redox conditions, mixing conditions, pH, competitor ions, and sediment concentration (McCallister & Logan, 1978; Golterman, 2004; Withers & Jarvie, 2008). In rivers where PP levels are elevated, P adsorption/desorption to or from suspended and river bed sediment can be significant (Stone *et al.*, 1991; Petticrew *et al.*, 2006; Sharpley *et al.*, 2009; Kleinman *et al.*, 2010; Meals *et al.*, 2010) and result in significant and sustained ecological responses in stream biota (Silins *et al.*, 2014). While the sorption behavior of suspended and bed sediment in agriculturally impacted streams has been previously reported (Stone & Mudroch, 1989; Jarvie *et al.*, 2005), the P sorption characteristics of

suspended sediment in rivers draining severely burned forested landscapes are also largely unknown.

Given that the drinking water for at least two-thirds of American water supplies (Stein & Butler, 2004) and at least one-third of the world's largest cities, including Tokyo, Singapore, Melbourne, Los Angeles, and Rio de Janeiro (Dudley & Stolton, 2003), originates from forested watersheds, wildfire-associated increases in P inputs to high quality, typically oligotrophic streams are concerning because P is generally understood to be the limiting nutrient for algal and cyanobacterial growth in freshwater systems – elevated levels of these microorganisms can disrupt ecosystem health and integrity (Schindler, 1977). They can also result in elevated hepatotoxin and neurotoxin levels (e.g. microcystin, anatoxin) (Kotak *et al.*, 2000; Giani *et al.*, 2005). Accordingly, the potential changes in water quality that may be expected in conjunction with increased levels of PP may have significant infrastructure and cost implications for drinking water supply and treatment (Emelko *et al.*, 2011; Bladon *et al.*, 2014). Moreover, because P often limits aquatic system productivity (Elwood *et al.*, 1981), an understanding of its forms and mobility is essential for mechanistically linking physical/geochemical conditions and ecology in aquatic systems.

Given the potential implications of wildfire-associated P legacy effects on aquatic ecosystem health, and water quality and supply, the objectives of this investigation were to (1) characterize the spatial distribution of PP forms (NAIP, AP, OP) and phosphorus sorption characteristics (EPC_0) of suspended sediment to evaluate both the downstream propagation and longevity of wildfire effects on these parameters across a range of watershed scales, up to the large river basin scale, and (2) identify key geochemical mechanism(s) causing these long-term/large-scale legacy watershed effects.

Materials and methods

Study area

In 2003, the Lost Creek wildfire burned a nearly contiguous area of 21 000 ha in the headwaters regions of the Castle and Crowsnest River basins in southern Alberta, Canada (Fig. 1). This particularly severe wildfire consumed virtually all organic matter on the forest floor and resulted in increased sediment (Silins *et al.*, 2009b), nitrogen (Bladon *et al.*, 2008), and phosphorus (Silins *et al.*, 2014) export to downstream reaches of these rivers. Prior to the wildfire, the study area was not significantly logged; however, postfire salvage logging did occur in two of the watersheds (Lyons and Byron) (Table 1).

Elevation in the study area ranges from 1100 to 3100 m. The dominant forest species at lower elevations include lodgepole

pine (*Pinus contorta* var. *latifolia*), while Engelmann spruce (*Picea engelmannii*) and subalpine fir (*Abies lasiocarpa*) are dominant at higher elevations. Composition of the near stream vegetation is comparable to that of the contiguous, upland forest vegetation. Alpine ecozones include bare rock and alpine meadow vegetation (above the tree line). The geology in the study area is characterized by Cretaceous shales, limestone, dolomite, and sandstone overlain by well to imperfectly drained Eutric or Dystric Brunisol soils (Bladon *et al.*, 2008). Surface lithology is poorly connected to bedrock geology because of vast deposits of calcareous glacial moraines, till blankets, and till veneers formed during the late Quaternary Period. Because of the high slope and abundance of glacial-fluvial, erodible deposits (i.e. sands and gravels), gravel-bed rivers are predominant in the study area.

Mean annual precipitation in the headwaters region of the Oldman River ranges from 700 to 1700 mm yr⁻¹. Approximately 55% of the annual precipitation is rain during the frost free period. The mean annual temperature is 4.6 °C, and mean monthly temperatures vary between -7 °C in the winter and 16 °C in the summer. In 2009, the annual discharge of the Crowsnest and Castle Rivers was 249 and 430 mm yr⁻¹, respectively, whereas high snowpacks during the winter of 2009/2010 resulted in greater discharges in both rivers in 2010 (341 and 579 mm yr⁻¹, respectively). The hydrology of both rivers is snowmelt-dominated, peaking annually between May and early June; the highest stream flows are typically observed with late spring rain on snow events, or large convective or frontal storms. Notably, mean daily flows for the Crowsnest River [gauged by Water Survey of Canada at Frank, AB (Stn. No. 05AA008)] reflect a comparatively more stable flow regime than in the Castle River [gauged near Beaver Mines, AB (Stn. No. 05AA002)]. While the Castle River has experienced a comparatively greater proportion of very low flows, notably, it has also experienced a much greater proportion of higher flows (>0.75 mm d⁻¹) since the wildfire (2004–2010; Fig. 2). Detailed study site characteristics are presented in Table 1.

Suspended sediment geochemical sampling

Phillips samplers are commonly used to sample fine-grained suspended sediments (<100 µm in size) for the characterization of sediment-associated pollutants (Phillips *et al.*, 2000; Russell *et al.*, 2001; Ballantine *et al.*, 2008; Walling *et al.*, 2008; Collins *et al.*, 2010). They are generally deployed for extended periods to enable generally flow-weighted collection of sufficient quantities of fine sediment without significantly altering sediment properties (e.g. particle size distribution) (Phillips *et al.*, 2000). Although longer deployment periods often preclude numerous sequential samples from being collected at a given location, Phillips samplers enable robust characterization of terrestrial and lotic sediments because they integrate the chemical signatures of upstream sediment exports over relatively long time scales (e.g. months).

In the present investigation, suspended sediment was continuously sampled during ice-free conditions over a two-year

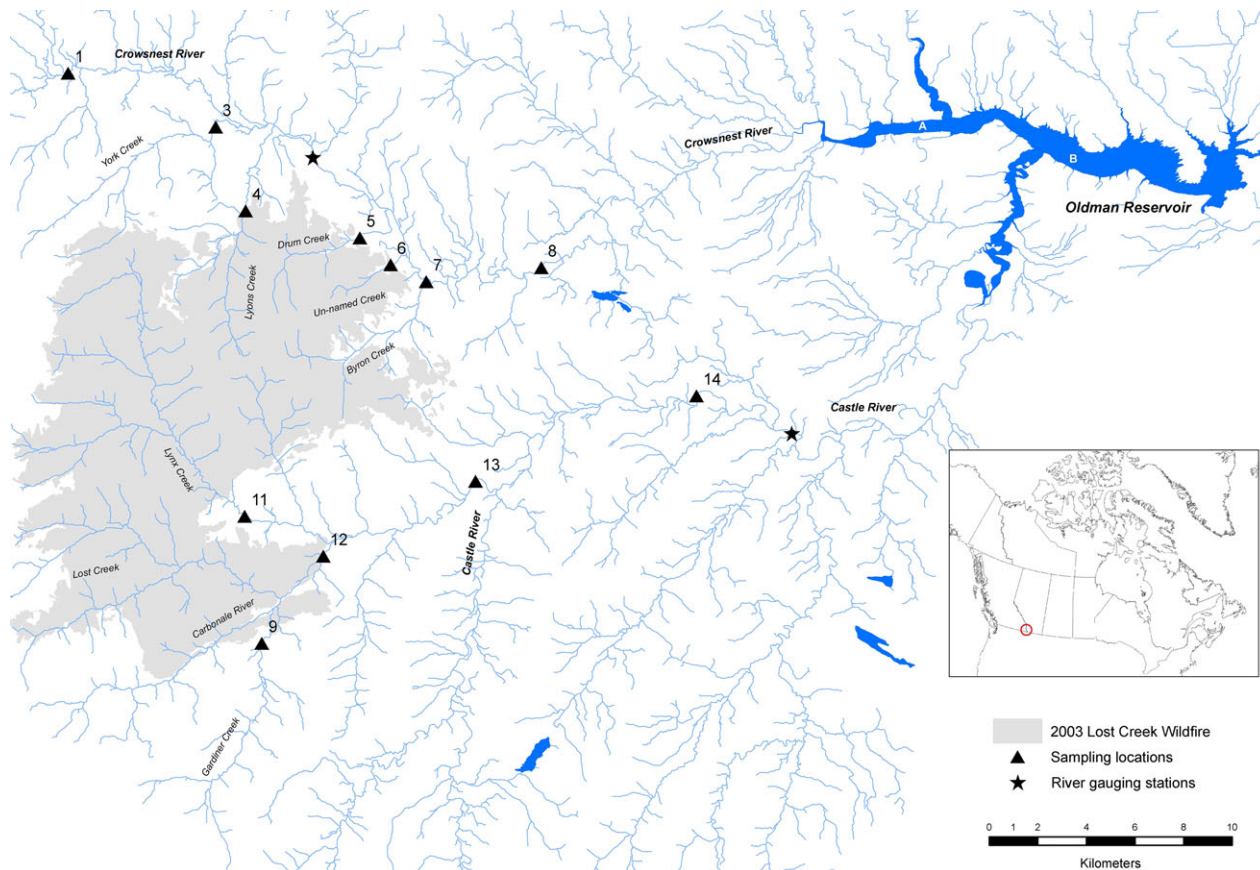


Fig. 1 The 2003 Lost Creek wildfire and sampling/gauging stations in the Crownsnest and Castle River watersheds. Sampling locations 1 and 9 are unburned upstream reference (R) locations in Crownsnest and Castle Rivers, respectively. Sampling locations 3 to 7 and 11 to 13 are within burned tributaries (BT), and locations 8 and 13, 14 are situated downstream of the burned tributary inputs (DBT) to the Crownsnest and Castle Rivers, respectively. A and B in the Oldman reservoir show location of Ponar sample locations (Fig. 5).

period. Phillips samplers were deployed at 14 locations across two watersheds (Fig. 1, Table 1) to provide sufficient quantities of sediment to assess the geochemical signatures of all upstream sources. Sediment was collected during five consecutive periods (2009 April to July, July to August, and August to October; 2010 April to August, and August to October). The samplers were placed at 0.6 times the stream depth in the centroid of flow. Sampling depth was periodically adjusted as flow conditions varied throughout the season. At the end of each sampling period, the samplers were emptied into acid-washed triple-rinsed plastic pails, then cleaned and immediately redeployed. After 48 h, excess water was decanted from the pails. The sediment samples were stored in coolers at 4 °C during shipment and subsequently freeze-dried for further analyses.

Sampling locations were chosen to reflect both burned, upstream watersheds in the headwaters and larger watersheds well downstream of the wildfire, including (1) upstream reference (unburned) watersheds, (2) upstream tributary watersheds impacted by the wildfire, and (3) locations downstream of burned tributary inputs on the larger Crownsnest and Castle River systems (Table 1). Sites 1 and 9 are in the

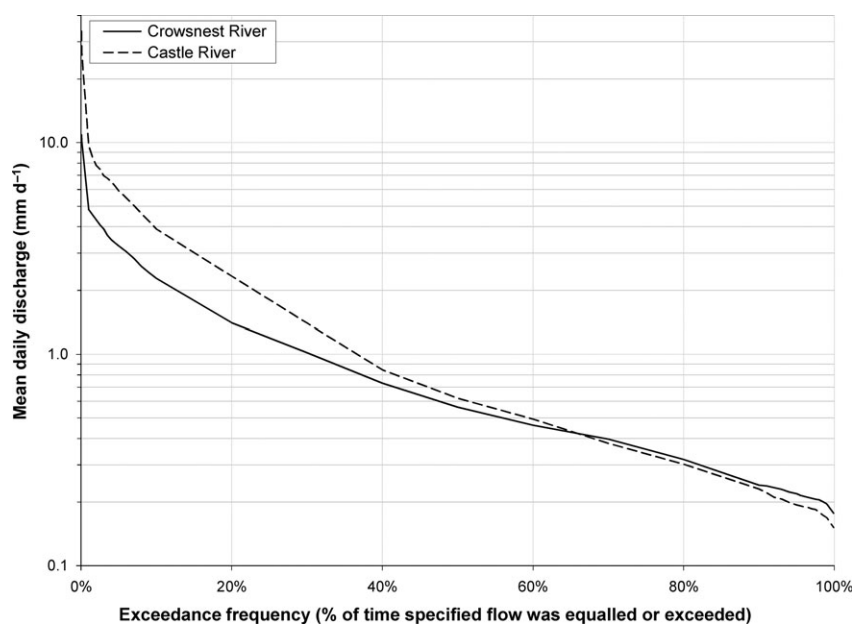
Crownsnest and Castle River systems, respectively, and drain predominantly forested, reference (unburned) landscapes upstream of the area burned by the Lost Creek wildfire. Sites 3–7 and 11–13 drain streams in wildfire-impacted tributaries that flow directly into the Crownsnest and Castle Rivers, respectively (Fig. 1). Sites 8 and 14 are located downstream of burned tributary inputs to the Crownsnest and Castle Rivers, respectively.

Physical characteristics and major element composition of sediment

Sediment physical characteristics and elemental composition were analyzed at a commercial laboratory (Act Labs, Burlington, ON, Canada) according to standard methods. The median diameter (D_{50}) and specific surface area (SSA) of the collected sediments were measured with a Malvern Mastersizer 2000. Estimation of SSA assumed spherical particles. Major elements (Al, Fe, Mg, Ca, Mn) were evaluated by X-ray fluorescence, and the results were reported as percent dry weight (Mudroch, 1985). Analytical accuracy was confirmed using Canadian Reference Standards

Table 1 Characteristics of the study sites

Site	River	Upstream drainage area (ha)	Upstream burn area (ha)	% Upstream burned	% Upstream salvage logged
Crowsnest River					
Reference (R)					
1	Crowsnest River	16 076	0	0	0
Burned Tributaries (BT)					
3	York Creek	3365	271	8	0
4	Lyons Creek	2650	1703	64	21
5	Drum Creek	1179	1064	90	0
6	Unnamed Creek	478	450	94	0
7	Byron Creek	2511	1508	60	5
Downstream (DBT)					
8	Crowsnest River	55 387	5300	10	1
Castle River					
Reference (R)					
9	Gardiner Creek	3646	0	0	0
Burned Tributaries (BT)					
11	Lynx Creek	7369	6113	83	0
12	Carbondale River	15 050	5781	38	0
13	Carbondale River	31 234	13 867	44	0
Downstream (DBT)					
14	Castle River	79 749	14 167	18	0

**Fig. 2** Flow regime (area normalized mean daily flow) of the Crowsnest and Castle Rivers for a 7-year period after the Lost Ck. wildfire (2004–2010).

AGV-1, MRG-1, NCM-N, GSP-1, and SY-3. Sediment total organic carbon (i.e. particulate organic carbon; POC) was determined using solid state infrared absorption (Eltra CS-2000 Carbon/Sulfur Analyzer; ELTRA GmbH, Haan, Germany); the results were accurate to 1%.

Fractionation of phosphorus forms

A sequential extraction scheme was used to fractionate PP into five operationally defined P fractions (Boström *et al.*, 1982; Pettersson *et al.*, 1988; Stone & English, 1993). Nonapatite

P (NAIP) is defined as the sum of three reactive phosphate fractions: loosely sorbed P (1.0 M NH_4Cl -P extractable P), reductant soluble P (0.11 M $\text{NaHCO}_3\cdot\text{Na}_2\text{S}_2\text{O}_4$ extractable P), and metal oxide bound P (1.0 M NaOH extractable P) (Boström *et al.*, 1982). The loosely sorbed P fraction is generally considered a seasonally variable pool of P compounds dissolved in sediment interstitial water (Rydin, 2000; Kaiserli *et al.*, 2002). It includes porewater P, P released from CaCO_3 -associated complexes, and P leached from decaying biomass (Gonsiorczyk *et al.*, 1999; Pettersson, 2001; Kaiserli *et al.*, 2002). The reductant soluble P fraction is considered as a potentially mobile P pool that is available for primary production (Kleeberg & Dudel, 1997; Kaiserli *et al.*, 2002). This fraction includes redox sensitive P forms (typically associated with Fe hydroxides and Mn compounds) (Kozerski & Kleeberg, 1998; Kaiserli *et al.*, 2002) that can act as internal/in-stream sources of P when sediments are anaerobic due to dissolved oxygen depletion (Kleeberg & Dudel, 1997; Kaiserli *et al.*, 2002). The metal oxide bound P fraction is a measure of P that is also available for primary production (Zhou *et al.*, 2001; Kaiserli *et al.*, 2002). It is often used for assessing the short- and long-term availability of P in sediments because it represents P bound to metal oxides (mainly Fe and Al) that can be released and contribute to primary productivity when anoxic or aerobic conditions prevail at the sediment-water interface (Ting & Appan, 1996; Kaiserli *et al.*, 2002).

Apatite P is the 0.5 M HCl extractable P fraction bound primarily to Ca and Mg carbonates and therefore represents P forms that are sensitive to low pH. This fraction is assumed to consist mainly of apatite P (natural and detrital), carbonate bound P, and traces of hydrolysable organic P. Thus, it is considered a relatively stable, permanent sink of P in sediments (Kozerski & Kleeberg, 1998; Gonsiorczyk *et al.*, 1999; Kaiserli *et al.*, 2002).

The organic P (OP) fraction is extracted using hot 1 M NaOH (85 °C). While inorganic forms of phosphate, such as HPO_4^{2-} and H_2PO_4^- , have been the focus of many investigations because they predominate biologically available P forms in sediments, OP composition and bioavailability in sediments remain relatively poorly understood. Particulate OP forms include a range of living and detrital organic molecules such as nucleic acids, phospholipids, phosphomonoesters and diesters, phosphonates, and nucleotides (Worsfold *et al.*, 2008).

After each extraction and subsequent centrifugation, supernatants were analyzed for molybdenum reactive P using a Technicon™ Autoanalyzer II (SEAL ANALYTICAL, Mequon, WI, USA) according to the ammonium molybdate/stannous chloride method (Environment Canada, 1979). At least 10% of the extractions were performed in triplicate. The mean coefficient of variation (CV) was <6 % for all PP forms (NAIP, AP, and OP) analyzed.

Phosphorus adsorption/desorption potential

The equilibrium phosphorus concentration (EPC_0) is a measure of the potential of sediments to adsorb or release SRP depending on the ambient SRP concentrations in aquatic

systems (House & Denison, 1998, 2000). The EPC_0 is determined experimentally by plotting the measured mass of P sorbed per mass of sediment vs. the initial concentration of SRP prior to contact with the sediment (Taylor and Kunishi, 1971; Froelich, 1988). It can be used to estimate P flux and transfers in aquatic systems (House *et al.*, 1995). To estimate the adsorption/desorption potential of SRP from suspended sediment to the water column, batch equilibrium experiments were conducted to determine the EPC_0 of suspended sediments collected at the sampling sites detailed in Table 1. A six-point isotherm was established by mixing 0.25 g of freeze-dried sediment with 25 mL of background water matrix (0, 10, 25, 50, 100, 200 $\mu\text{g L}^{-1}$ KH_2PO_4) in 50-mL polypropylene centrifuge tubes. The triplicate sorption experiments were conducted for 18 h at room temperature of 24 ± 1 °C, on an orbital shaker table set to low speed (~50 rpm). The tubes were then centrifuged (4000 g for 5 min), and the supernatant was filtered using 0.45- μm polypropylene syringe filters. SRP in the supernatant was analyzed using the ammonium molybdate/stannous chloride method (Environment Canada, 1979).

Statistical analyses

Representative characterization of sediment geochemical properties in upstream and downstream reaches of the Castle and Crowsnest Rivers was achieved by continuous sampling of suspended solids over a range of hydrologic flow conditions (baseflow, snowmelt freshet, stormflows) during the two-year (2009–2010) study period. Differences between PP forms in upstream and downstream reaches of the rivers and their wildfire-impacted tributary inputs were compared using an 'independent samples *t*-test' (SPSS 21, IBM Corp., 2012). The P speciation data met the assumptions of normality according to Q-Q and P-P plots, as well as the Shapiro-Wilk test of normality. Homogeneity of variances was confirmed using Levene's test. Differences in the EPC_0 among sampling locations were evaluated by least squares linear regression of P mass sorbed per mass of sediment on initial SRP concentration and comparison of regression slopes and intercepts using modified *t*-tests (after Zar, 1999).

In this investigation, an ~10% significance level ($\alpha = \sim 0.1$) was considered strong evidence of significant effects. This significance level is appropriate because the study systems are dynamic – hydro-climatic conditions vary considerably at larger river basin scales such as those investigated herein. Accordingly, the longevity and spatial extent of wildfire impacts on water quality and associated ecological recoveries also can vary considerably at these scales. Undoubtedly, the greatest effects of wildfire in the Crowsnest and Castle River systems did not occur either concurrently or exactly 6–7 years after the wildfire when the investigation was conducted. While a more stringent significance level (e.g. $\alpha = 0.05$) could be applied, it would result in the same general inferences regarding sediment-phosphorus dynamics after wildfire, but a less complete picture of key mechanisms regulating downstream P legacy effects of wildfire.

Results

Sediment characteristics and elemental content

The particle size characteristics and elemental content of suspended sediments collected in the Crowsnest and Castle Rivers are presented in Table 2. A significant fraction of the sediment mass in the study streams was fine grained, thereby contributing significant surface area for contaminant and P adsorption/desorption. Specifically, the median sediment grain diameter (D_{50}) ranged from approximately 40 to 50 μm and specific surface area (SSA) ranged from approximately 0.5–1 $\text{m}^2 \text{g}^{-1}$.

Concentrations of Al, Fe, Mn, and Ca were significantly higher downstream of the wildfire (Site 8) in the Crowsnest River, compared to the upstream reference Site 1 ($P \leq 0.065$ in all cases; details in Table 3), whereas Mg was significantly lower ($P \leq 0.001$; Table 3). Significant differences in some major elements and POC were not observed between upstream reference Site 1 and downstream Site 14 in the Castle River; the only exceptions were Mn and Ca, which significantly increased ($P = 0.078$ and $P = 0.006$, respectively; Table 3) and Mg, which significantly decreased ($P = 0.009$; Table 3) downstream of the wildfire.

Concentrations of Al, Fe, Mn, and POC were significantly higher in wildfire-impacted tributaries of the Crowsnest River, compared to upstream reference Site 1 ($P \leq 0.094$ in all cases; details in Table 3). In contrast, Ca and Mg were significantly lower ($P \leq 0.001$; Table 3). Significant differences in major elements and

POC were not observed between upstream reference Site 1 and the burned tributaries of the Castle River; the only exception was Ca, which significantly increased in the burned tributaries ($P = 0.001$).

Concentrations of Ca were significantly higher downstream of the wildfire (Site 8) in the Crowsnest River, compared to upstream locations in the burned tributaries at Sites 3–7 ($P \leq 0.001$; Table 3). Here, Al, Fe, and POC were significantly lower ($P \leq 0.089$ in all cases; details in Table 3). Significant differences in major elements and POC between upstream reference Site 14 and upstream locations in the burned tributaries (Sites 11–13) in the Castle River were only observed in elevated Al ($P = 0.072$) and decreased Ca ($P = 0.052$) (Table 3).

Total and fractional composition of particulate phosphorus

The concentrations of PP forms (TPP, NAIP, AP, OP) at the sampling sites in the Crowsnest and Castle Rivers are presented in Fig. 3. The mean and standard deviation of the concentrations of TPP and the five sequentially extracted PP fractions [NAIP (P1:loosely sorbed, P2:reductant soluble, and P3:metal oxide bound P), AP and OP] measured in the rivers are presented in Table 4.

Total particulate phosphorus increased downstream of wildfire-impacted areas in both study rivers. It was significantly higher downstream of the wildfire (Site 8) in the Crowsnest River, compared to both upstream reference Site 1 ($P = 0.001$) and the wildfire-impacted

Table 2 Particle size characteristics and elemental composition of Crowsnest and Castle River sediments [mean and standard deviation (SD)]

River	Site	<i>n</i>	D_{50}	SSA	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Particulate organic carbon (%)
			(μm)	($\text{m}^2 \text{g}^{-1}$)	(%)	(%)	(%)	(%)	(%)	
			Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)
Crowsnest	Upstream reference 1	5	40.4 (20.3)	0.9 (0.8)	8.3 (0.7)	2.7 (0.2)	0.1 (0.0)	2.2 (0.1)	9.8 (0.5)	4.8 (1.0)
	Burned tributaries 3–7	23	41.3 (23.2)	0.9 (0.6)	11.0 (0.7)	4.0 (0.6)	0.1 (0.1)	1.7 (0.3)	4.3 (1.0)	6.2 (2.3)
	Downstream of burned tributaries 8	5	45.5 (25.9)	0.8 (1.0)	8.9 (0.5)	3.3 (0.2)	0.1 (0.0)	1.6 (0.2)	12.3 (1.8)	4.8 (0.8)
Castle	Upstream reference 9	4	37.7 (26.2)	1.1 (1.0)	11.2 (0.7)	3.9 (0.3)	0.1 (0.0)	3.2 (0.1)	2.1 (0.6)	2.8 (2.5)
	Burned tributaries 10–13	13	51.0 (42.2)	0.8 (0.7)	10.3 (1.4)	3.7 (0.5)	0.1 (0.0)	3.2 (1.1)	6.0 (3.5)	3.1 (1.3)
	Downstream of burned tributaries 14	2	49.4 (16.0)	0.5 (0.2)	11.0 (0.0)	4.0 (0.2)	0.1 (0.0)	2.7 (0.2)	4.3 (0.1)	3.0 (0.2)

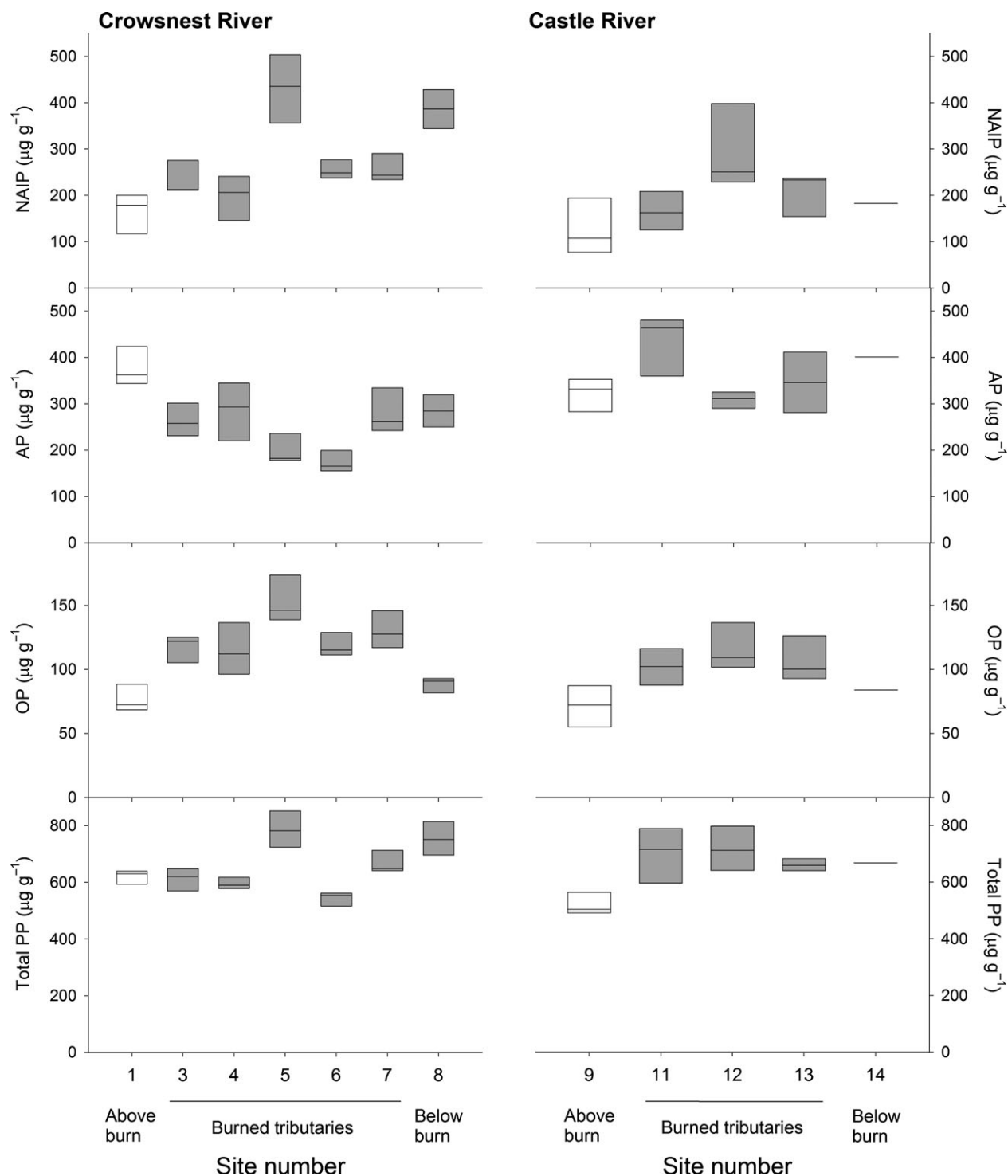


Fig. 3 Distribution (median, upper, and lower quartiles) of particulate phosphorus (PP) forms [nonapatite inorganic phosphorus (NAIP), apatite phosphorus (AP), organic phosphorus (OP), Total particulate phosphorus (TPP)] in the Crowsnest and Castle Rivers for unburned upstream reference (R) locations, locations within burned tributaries (BT), and downstream of the burned tributary inputs (DBT) for 2009/2010.

AP was significantly higher in the wildfire-impacted tributaries (Sites 11–13), compared to the upstream reference Site 9 ($P = 0.089$; Table 3).

Organic phosphorus comprised the smallest fraction of TPP at all the sampling sites in both the Crowsnest and Castle Rivers (Table 4). Sediment OP was

Table 4 Total particulate phosphorus (TPP) and P fractions in suspended sediment of the Crowsnest and Castle Rivers (mean and standard deviation). P1 – loosely sorbed P; P2 – reductant soluble P; P3 – metal oxide bound P; P4 – apatite P; and P5 – organic P

River	Site	<i>n</i>	Nonapatite inorganic phosphorus			Apatite phosphorus	Organic phosphorus	TPP ($\mu\text{g P g}^{-1}$) Mean (SD)
			P1 ($\mu\text{g P g}^{-1}$) Mean (SD)	P2 ($\mu\text{g P g}^{-1}$) Mean (SD)	P3 ($\mu\text{g P g}^{-1}$) Mean (SD)	P4 ($\mu\text{g P g}^{-1}$) Mean (SD)	P5 ($\mu\text{g P g}^{-1}$) Mean (SD)	
Crowsnest	Upstream reference 1	5	23.2 (10.6)	64.3 (15.9)	75.1 (26.1)	379.3 (42.0)	77.2 (13.2)	619.2 (24.1)
	Burned tributaries 3–7	23	26.1 (12.2)	97.2 (54.9)	152.6 (42.8)	248.0 (62.4)	128.0 (23.7)	651.9 (94.0)
	Downstream of burn 8	5	67.9 (24.5)	181.2 (45.2)	137.3 (11.1)	284.4 (35.0)	87.9 (6.0)	758.7 (67.7)
Castle	Upstream reference 9	4	19.8 (24.8)	35.6 (32.1)	70.7 (16.2)	322.2 (37.9)	71.5 (16.8)	519.7 (42.1)
	Burned tributaries 10–13	13	26.9 (23.3)	69.2 (47.7)	98.3 (30.1)	374.1 (69.3)	98.6 (25.7)	667.0 (96.2)
	Downstream of burn 14	2	21.8 (13.7)	76.0 (23.6)	85.0 (10.1)	401.2 (121.1)	83.8 (2.6)	667.7 (103.8)

significantly higher in both wildfire-impacted tributaries (Sites 3–7) and downstream of the wildfire (Site 8) in the Crowsnest River, compared to the upstream reference Site 1 ($P < 0.001$ and $P = 0.068$, respectively; Table 3). Here, OP significantly decreased between the wildfire-impacted tributaries (Sites 3–7) and the sampling location downstream of the wildfire (Site 8) ($P < 0.001$; Table 3). In the Castle River, sediment OP was significantly higher only in the wildfire-impacted tributaries (Sites 11–13), compared to the upstream reference Site 9 ($P = 0.035$; Table 3).

Phosphorus adsorption/desorption potential

The P adsorption/desorption profiles obtained during the batch experiments are presented in Fig. 4. This figure shows that the sediment EPC_0 generally increased between the upstream reference Sites (1 and 9) and the sampling locations downstream of the wildfire (Sites 8 and 14) in both the Crowsnest and Castle Rivers, respectively. Notably, the mean EPC_0 ($105.7 \mu\text{g P L}^{-1}$) increased by 39% in burned tributaries (Sites 3–7) relative to that ($76 \mu\text{g P L}^{-1}$) of upstream reference Site 1 in the Crowsnest River ($P < 0.001$). It further increased to $213.5 \mu\text{g P L}^{-1}$ (181% greater than unburned reference Site 1; $P < 0.001$) at the sampling location downstream of the wildfire (Site 8) (Fig. 4). Changes in EPC_0 in the Castle River were less pronounced downstream of the burned tributaries. Although the 39% increase in mean EPC_0 from burned tributaries (Sites 11–13; mean of $77.5 \mu\text{g P L}^{-1}$ compared to $55.6 \mu\text{g P L}^{-1}$ in reference Site 9; $P < 0.001$) was similar to that observed in the Crowsnest River, the mean EPC_0 at Site 14

downstream of the burned tributaries ($92.7 \mu\text{g P L}^{-1}$) was only 67% greater than in the unburned reference Site 9 ($P < 0.001$; Fig. 4).

Discussion

Despite this study being conducted 6 and 7 years after the Lost Creek wildfire, significant increases in sediment-associated P and bioavailable P fractions (NAIP) were still evident in a range of burned tributaries draining into both the Crowsnest and Castle Rivers. Moreover, the downstream propagation of these effects was still detectable at larger basin scales. While both river systems exhibited elevated TPP downstream of wildfire-impacted areas, wildfire-associated increases in NAIP, POC, and the EPC_0 , as well as some sediment-associated metals (e.g. Al, Fe, Mn, and Ca), were more pronounced in the Crowsnest River than the Castle River. River systems are dynamic; therefore, the contrast between the study systems is important to consider at broader temporal scales than the study period.

The absolute magnitude of flow ($\text{m}^3 \text{s}^{-1}$) in the Castle River is ~10 times that of the Crowsnest River. Comparatively higher flows and associated shear stresses that govern sediment erosion, transport, and deposition in the Castle River would result in fewer settling opportunities and greater scouring of deposited fine sediments from the channel. In this system, it is likely that a larger portion of the wildfire-associated fine sediments had already been transported downstream to, and deposited in, either floodplains or the Oldman Reservoir (Fig. 1) during the first five years after the wildfire.

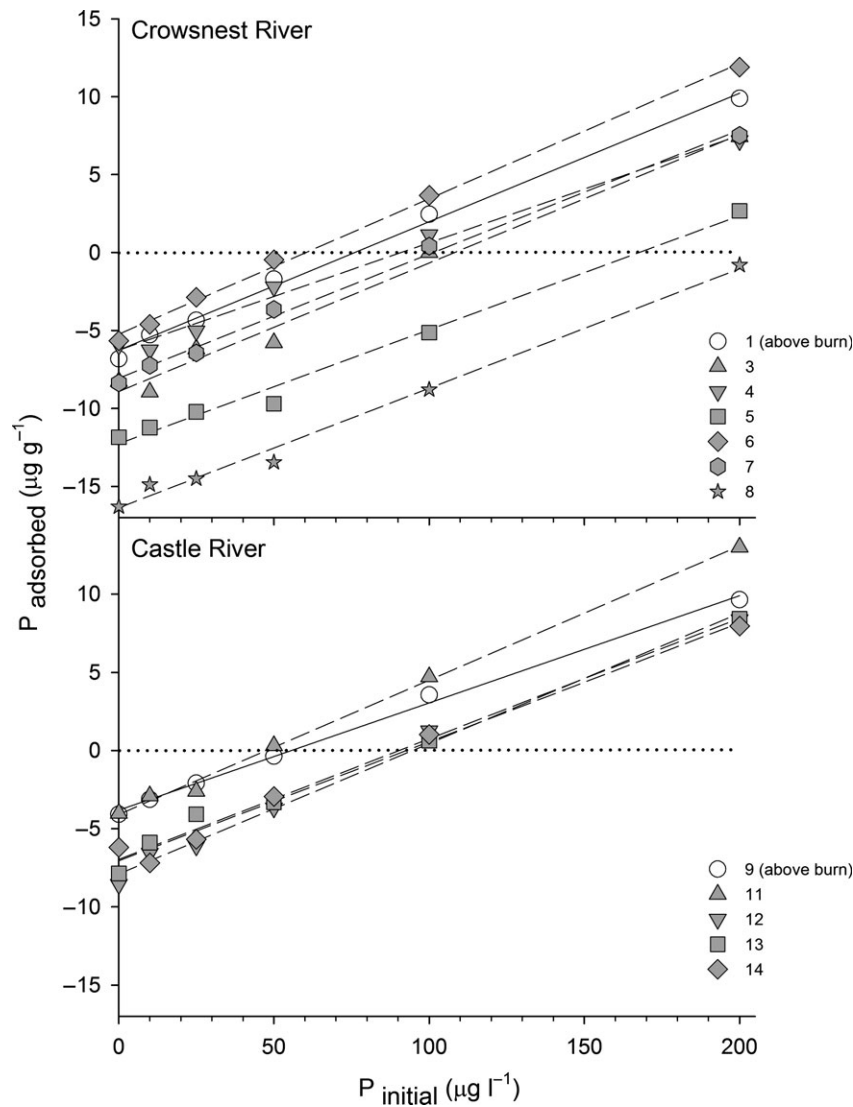


Fig. 4 Phosphorus sorption characteristics of suspended sediment collected from unburned upstream reference (R) locations (Sites 1, 9), burned tributary input (BT) locations (Sites 3–7 and 11–13), and downstream of the burned tributary input (DBT) locations (Sites 8, 14) in the Crowsnest and Castle Rivers, respectively. The EPC_0 for each site is the P concentration on the x axis corresponding to P adsorbed = 0 (dotted horizontal line). $R^2 > 0.99$ for all adsorption/desorption profiles.

Indeed, post-wildfire-associated sediment deposits were evident below the Castle River confluence with the reservoir (Figs 1 and 5a), whereas no clear evidence of wildfire inputs to the reservoir was observed in the western, Crowsnest River arm of the reservoir (Figs 1 and 5b). Accordingly, 'identical' signatures of wildfire impacts on sediment-associated TPP, P fractions, and P sorption 6 to 7 years after the wildfire in these neighboring systems would not necessarily be expected. Because of generally lower, more stable flows (Fig. 2), it would be expected that these postfire impacts would remain more pronounced over time in the Crowsnest River – this was generally observed. While both river

systems exhibited elevated TPP downstream of wildfire-impacted areas, wildfire-associated increases in NAIP, POC (Tables 3 and 4; Fig. 3), and the EPC_0 (Fig. 4), as well as some sediment-associated metals such as Al, Fe, Mn, and Ca (Tables 3 and 4), were more pronounced in the Crowsnest River, relative to the Castle River. Accordingly, subsequent discussion of wildfire impacts on sediment-associated P forms and sorption behavior predominantly focuses only on results from the Crowsnest River and its burned tributaries. It can be reasonably expected that similar behavior was exhibited in the Castle River at an earlier point in time after the wildfire, as suggested by still

noticeably elevated TPP, reductant soluble P, and Ca levels in this system (Tables 3 and 4).

Land use change can influence the nature and distribution of PP forms sequestered in bed sediment in river beds (McCallister & Logan, 1978; Stone & Mudroch, 1989; Stone & English, 1993; Fogal *et al.*, 1995) and suspended sediment (Logan *et al.*, 1979; De Pinto *et al.*, 1981). In this investigation, a significant fraction of the sediment mass in the study streams and rivers was fine grained (Table 2) and much of it was originally delivered through wildfire-enhanced terrestrial erosion and streambank mass wasting processes; >80% of the downstream sediment geochemical contributions in the wildfire-impacted study areas were produced by only 14% of the affected upstream landscape (Stone *et al.*, 2014). This predominance of wildfire derived, P-enriched fine sediment caused the observed increases in TPP and the associated increased levels of NAIP (Tables 3 and 4; Fig. 3), as well as metals such as Al and Fe (Tables 2 and 3) and reductant soluble (i.e. redox sensitive P

forms typically associated with Fe hydroxides and Mn compounds) and metal oxide bound sediment P (Tables 3 and 4). These results are consistent with published reports that indicate that metal levels can be generally elevated in both soils and stream sediments for some period after wildfire (Chambers & Attiwill, 1994; Parra *et al.*, 1996; Gallaher *et al.*, 2002; Ranalli & Stevens, 2004; White *et al.*, 2006; Biswas *et al.*, 2007; Silins *et al.*, 2009b; Witt *et al.*, 2009; Woodruff & Cannon, 2010; Emelko *et al.*, 2011; Writer & Murphy, 2012; Bladon *et al.*, 2014). In contrast, observed decreases in AP were likely linked to geological controls because the AP fraction most likely consists of Mg carbonates that are predominant in the calcareous till veneers in these basins. Notably, despite the length of time since the wildfire, NAIP in burned tributaries and its downstream transport still remained strongly associated with the proportion of watershed area burned across a broad range of smaller upstream, and larger downstream watershed scales in the Crowsnest River ($P < 0.034$). In contrast, these relationships were somewhat weaker ($P < 0.102$) in the Castle River basin, although arguably still notable and indicative of earlier wildfire impact (Fig. 6).

Particulate phosphorus forms in wildfire-impacted and reference sites in the present study were compared with some examples of previously reported data to illustrate the effect of different land disturbances (i.e. wildfire, agriculture, and urban intensification) on the relative range and concentration of PP forms reported in various rivers (Fig. 7). In this figure, the data from the two study river systems are composite mean values that include all sampling periods. Concentrations of TPP in the Crowsnest and Castle Rivers after wildfire were comparable to those observed in predominantly agricultural watersheds of the Maumee, Sandusky, and Cattaraugus Rivers in the United States (De Pinto *et al.*, 1981) where the use of inorganic and organic fertilizers contributes P to the land-water continuum; substantially higher TPP levels have been reported in agriculturally impacted rivers elsewhere (Walling *et al.*, 2001; Ballantine *et al.*, 2008). TPP levels in urbanized watersheds typically reflect P inputs from point sources that include sewage treatment works, storm tanks, and combined sewer overflows (Owens & Walling, 2002), and are also typically higher than those observed in agriculturally impacted watersheds, and wildfire-impacted watersheds, as demonstrated herein (Fig. 7). This analysis demonstrates that eutrophication occurs in other aquatic systems with TPP levels comparable to those observed in the wildfire-impacted rivers in the present investigation.

Ideally, a relatively simple metric such as NAIP would be directly indicative of increased primary

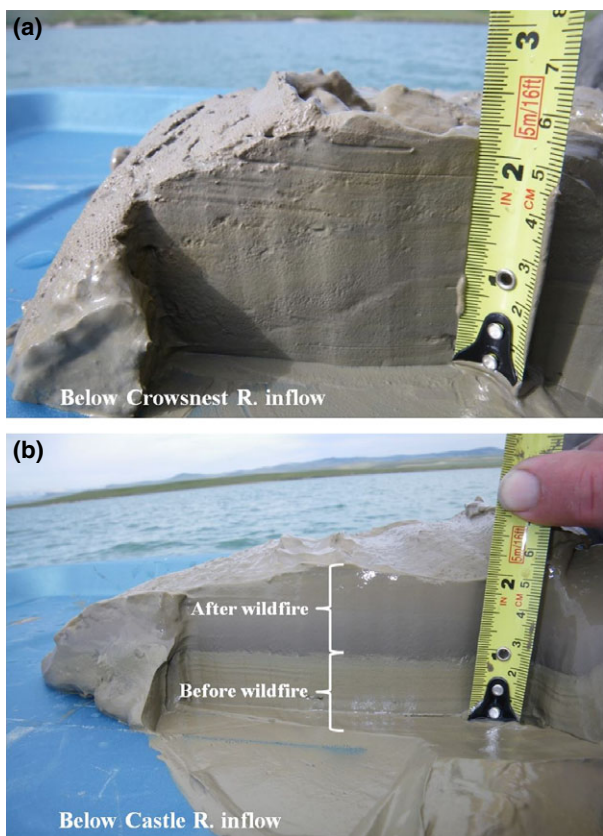


Fig. 5 Wildfire-associated fine sediment deposition (sampled using a Ponar sampler in August 2009) in the Oldman Reservoir. Reservoir sediments illustrated in photos (a) and (b) are typical of fine-grained reservoir sediments sampled below the Castle and Crowsnest River inflows, respectively (Photo credit: Southern Rockies Watershed Project).

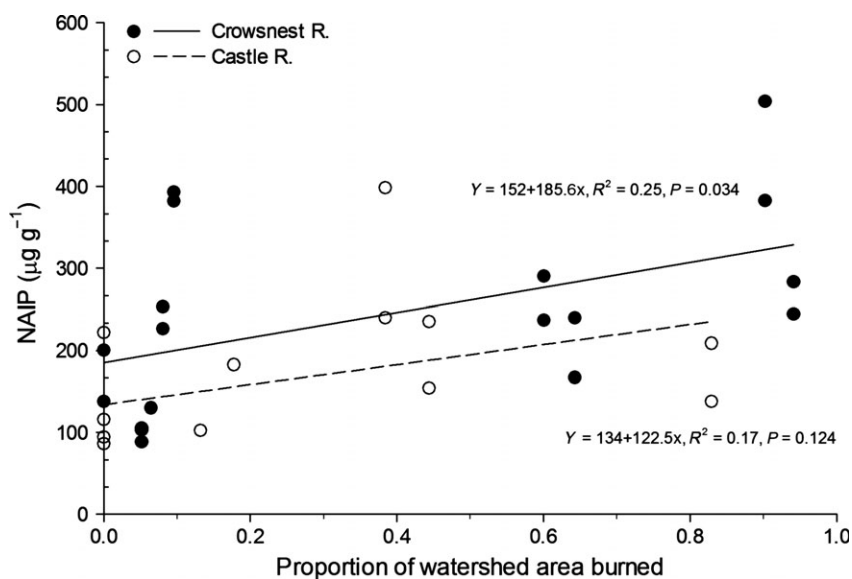


Fig. 6 Distribution of nonapatite inorganic phosphorus (NAIP) [most bioavailable form of particulate phosphorus (PP)] in the Crowsnest and Castle river basins as a function of proportion of watershed area burned.

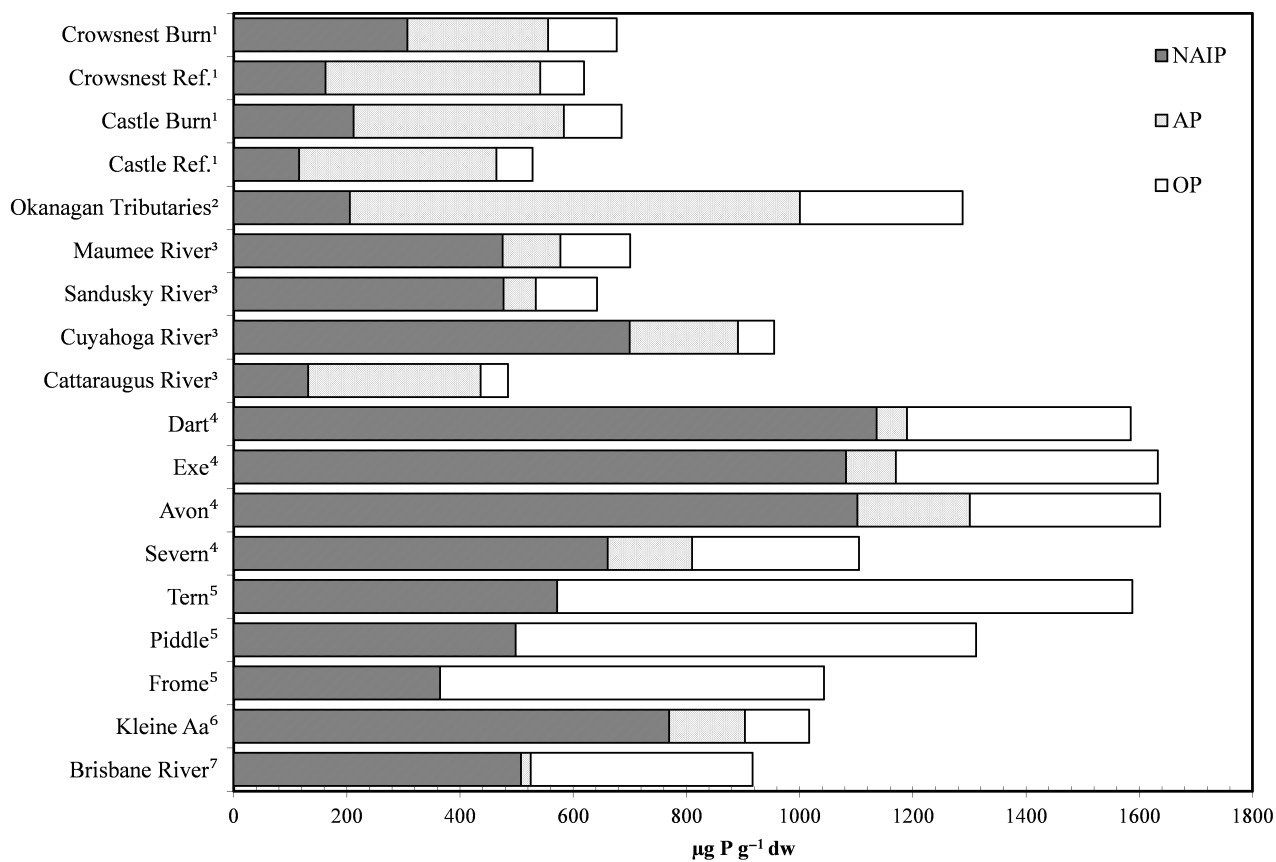


Fig. 7 Comparison of sediment-associated particulate phosphorus (PP) forms in the Crowsnest and Castle Rivers to those observed in agricultural and urbanized watersheds. The data from the two study river systems are composite mean values that include all sampling periods. Ref.: 1. This Study, 2. Gray & Kirkland (1986), 3. De Pinto *et al.* (1981) 4. Walling *et al.* (2001), 5. Ballantine *et al.* (2008), 6. Pacini & Gachter (1999), 7. Kerr *et al.* (2011).

productivity and eutrophication potential in aquatic systems; however, NAIP varies considerably across river systems (Fig. 7). Indeed, for eutrophication to occur, bioavailable P must be released to the water column; therefore, the relative relationship between EPC_0 (i.e. P release potential due to desorption from fine sediment) and dissolved P must be concurrently considered, because the nature and distribution of PP forms can significantly alter dissolved P (particularly SRP/orthophosphate) concentrations in aquatic systems (Froelich, 1988; Fox, 1993). Notably, unlike the 35–200 $\mu\text{g L}^{-1}$ SRP concentrations observed in the agriculturally impacted and urbanized rivers characterized in Fig. 7, streams draining relatively undisturbed forested regions of the eastern slopes of the Rocky Mountains in southern Alberta are typically oligotrophic; indeed, the mean concentration of SRP at the Crowsnest and Castle River reference locations (Sites 1 and 9) was $\sim 5 \mu\text{g L}^{-1}$ (Howery, 2010) and substantially lower than in those other rivers. At such low ambient SRP concentrations, substantial SRP release into the overlying water column would be expected from P-enriched fine sediment in the wildfire-impacted river systems (Fig. 4). As postfire impacts remained more pronounced in the Crowsnest River 6 and 7 years after the wildfire, more pronounced differences between wildfire-impacted and reference sediment EPC_0 in the Crowsnest River were also expected and observed (Fig. 4). If the wildfire-impacted sediment from the Crowsnest and Castle rivers was released to other river systems with higher ambient SRP concentrations (perhaps reflecting urban sources), the in-stream bioavailable P loading potential would likely be lower, and at higher SRP concentrations the fine-grained sediment would act as a P sink (Fig. 4).

The sediment-phosphorus dynamics described above can cause shifts in aquatic ecology after wildfire. P

often limits aquatic system productivity (Schindler, 1977; Elwood *et al.*, 1981). The combination of elevated P and greater light availability after loss of riparian canopy has been associated with increased algal production after wildfire (Minshall *et al.*, 2001). In addition to elevated levels of bioavailable NAIP (Tables 3 and 4; Fig. 3) that were expected to be released to the water column by desorption from postfire P-enriched fine sediment (Fig. 4), significant responses in stream algae (i.e. ~ 5 –71 times greater productivity; Fig. 8a) and associated algal/microbial river bed biostabilization (Stone *et al.*, 2011) also were observed in the study area following the Lost Creek wildfire. Increased algal production often leads to relative increases in benthic macroinvertebrate abundance (Earl & Blinn, 2003; Minshall, 2003; Spencer *et al.*, 2003; Silins *et al.*, 2014) and increased size of some aquatic predators. These cascading ecosystem impacts were observed in the study area where chlorophyll *a* concentrations ($P = 0.022$; Fig. 8a), macroinvertebrate abundance (Fig. 8b), and fresh weight of 2-year-old to 3-year-old cutthroat trout ($P = 0.014$; Fig. 8c) increased after the wildfire. Thus, hydro-chemical sediment-phosphorus dynamics can cause shifts in aquatic ecology and the structure of aquatic food webs after wildfire.

It is critical to consider why land disturbances such as wildfire can have long-term, legacy impacts on internal bioavailable P loading potential and thus, long-term ecological legacy impacts such as those demonstrated herein, 6 and 7 years after wildfire. As sediment is the primary vector of P transport in aquatic systems (Baker & Richards, 2002), associated increases in P export to downstream environments (Bayley *et al.*, 1992; Hauer & Spencer, 1998; Burke *et al.*, 2005; Lane *et al.*, 2008; Silins *et al.*, 2014) that are coupled with increased sediment erosion rates after wildfire are not surprising and have

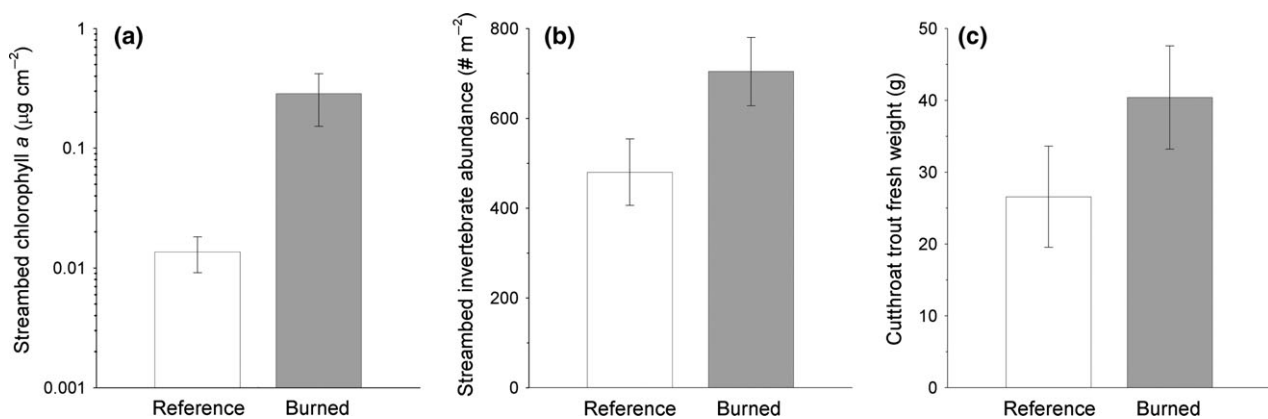


Fig. 8 Aquatic ecology of streams in the unburned (reference) and burned watersheds in the study area (modified from Silins *et al.*, 2014): (a) chlorophyll *a* concentrations (Chl*a*; $\mu\text{g cm}^{-2}$), (b) macroinvertebrate abundance [number of individuals per Surber sample (0.09 m^2)], and (c) fresh weight of 2-year-old to 3-year-old cutthroat trout (*Oncorhynchus clarkii*).

been commonly reported (Kunze & Stednick, 2006; Moody *et al.*, 2013), including in the present study watersheds (Silins *et al.*, 2009a). Notably, in the present investigation, a significant fraction of the sediment mass in the study streams was fine grained, thereby contributing significant surface area for P adsorption/desorption (Table 2) as well as increased opportunity for downstream transport due to lower settling velocities, which are associated with changes in sediment floc size, density, and porosity after the wildfire (Stone *et al.*, 2011). Collectively, the interaction of these components governs the longevity of watershed scale effects on P dynamics and aquatic ecology, as well as their disturbance-response trajectories. Such changes are ecosystem specific because of spatial and temporal variability in hydro-climatic conditions, geology, soils, topography, vegetation, river and bed morphology, and intensity and severity of disturbances such as wildfire, as well as proportion of watershed area impacted. Here, it should be underscored that both the Crowsnest and Castle Rivers, as well as their tributaries, have gravel beds that can entrap postfire, P-enriched fine sediment originating from hill slope runoff and bank erosion (Krishnappan & Engel, 2004; Glasbergen *et al.*, 2014). Accordingly, the release of SRP from fine interstitial sediment stored in these gravel beds is the mechanism by which bioavailable P is released to the water column both in headwaters and downstream reaches, across increasingly larger river basin scales where the importance of bed sediment storage will be more pronounced with increasing channel bed area. This same P legacy impact of wildfire would not be expected in other river systems where the potential for sediment entrapment is lower, or where bioavailable P enrichment is less pronounced despite the ingress of interstitial sediment.

Further research conducted at smaller spatial and temporal scales is required to rigorously evaluate P fluxes from fine grained sediment stored in gravel beds to the water column to better elucidate (1) river conditions that result in significant sediment entrapment, (2) P release dynamics from sediments sequestered in river beds, and (3) the relationship between P release and the presence of other nutrients in the development of phytomicrobenthos in gravel-bed river systems vulnerable to long-term legacy effects of land disturbance. Moreover, in these types of river systems, the combined effect of elevated NAIP and higher P release potential (EPC₀) from fine sediments such as those after wildfire may result in deteriorated water quality in downstream reservoirs – these changes can result in treatment challenges for drinking water supply, with significant implications for the infrastructure and operational costs of drinking water treatment (Emelko *et al.*, 2011).

Acknowledgements

The research was funded by the Natural Sciences and Engineering Research Council (NSERC) of Canada, Alberta Environment and Sustainable Resource Development (Forest Management Division), and Alberta Water Research Institute/Alberta Innovates – Energy and Environment Solutions. The assistance of Jeremy Fitzpatrick, Eddie Noton, Jolene Lust, and Jeremy Morrison with field work is gratefully acknowledged.

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