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Multi-Scale Variability of Heavy Metals in a Snowpack in the Triple Divide Region of the Western United States

Kelsey Hefner
Portland State University

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By

Kelsey Hefner

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Thesis Advisor
Kelly Gleason, Ph.D.

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ABSTRACT

Natural and anthropogenically sourced particulates are deposited from the atmosphere to landscapes via dry and wet deposition, making frozen winter snowpack a natural archive of atmospheric elemental composition. In the Western United States, wildfires are increasing in extent, duration, and severity. Severe fires remove forest canopy, impacting how atmospheric elements are dispersed and stored across snow-dominated watersheds. We evaluated concentrations of twelve elements in 397 winter snow core samples from a chronosequence of eight forests that burned with mixed severity from 2000 to 2018 in the Triple Divide region of Western Wyoming, the headwaters of the Columbia, Colorado, and Missouri Rivers. We detected the highest concentrations of Al, V, Cr As, and Pb in one fire scar south of Jackson Hole (p-values <0.05). We compared concentrations of all elements by forest structure classified into three forest types: unburned forests, burned forests, and open meadows. Concentrations of Al, Mn, Cr, Pb, V, and As in unburned forests were higher, and in some cases double that of burned forests and open meadows, (p-value < 0.05) likely due to forest canopy turbulence effects.

INTRODUCTION

1.1 Metals in the environment

Atmospheric metals are transported from the atmosphere and deposited to aquatic and terrestrial systems via wet and dry deposition processes, leading to their accumulation in remote environments.[1] Snow serves as an indicator of atmospheric constituents because of its ability to effectively scour and store atmospheric aerosols and particles, thereby acting as a natural archive of atmospheric deposition.[1–4] Trace elements encompass a range of metals and metalloids, and while metals occur naturally in Earth's crust and environment, atmospheric pulses of some toxic elements are linked to anthropogenic activities like fossil fuel and oil combustion, smelting, mining, manufacturing, waste incineration, agricultural fertilizer, cement production, residential wood combustion, and tire and break wear.[5–8]

The harmful effects of excess metals on ecosystems and human health have been well-documented. Excessive quantities of metals can be toxic to plants, invertebrates, fish, and wildlife, highlighting the importance of monitoring metal pollution in ecosystems.[9] [8,10,11]. Additionally, atmospheric transport is a dominant pathway for metals to reach remote places.[12] Biomagnification of toxic metals like Pb, Cd, Mo, V, and As through different trophic levels means that even low levels may lead to significant concentrations in plants and animals,[13] but monitoring these effects can be challenging due to the inherent logistic limitations in remote places. This paper aims to provide insight into the variability of metals within the subalpine region of the northern Rocky Mountain range to better understand the potential impacts of atmospheric metal deposition on remote ecosystems.

1.2 Wildfires alter forest structure

Currently, it is not well understood how atmospheric metals accumulate in snowpack in remote regions and across forest types with the introduction of wildfire disturbance. Globally, wildfires are changing ecosystems as they increase in extent, duration and severity.[14] Severe wildfires alter forest structure by removing forest canopy, exposing a snowpack to more solar radiation, higher temperatures, and greater wind velocities. [15–17] Wind-exposed snowpacks can either gain or lose mass and chemical load due to redistribution and wind-scouring.[16] In comparison, the presence of forest canopy can decrease snow accumulation via interception and loss by sublimation, or it can increase accumulation by protecting the snow's surface from incoming solar radiation and wind. The complex interactions between forest structure, snow accumulation, and atmospheric deposition highlight the need for further research to better understand the impact of wildfires on the cycling and fate of metals in the ecosystem.

To evaluate the influence of regional and forest structural changes on the concentrations of metals in snowpack, we measured elements Al, V, Cr, Mn, Ni, Cu, Zn, As, Se, Mo, Cd, and Pb an 18-year chronosequence of eight forest fires in the Triple Divide region of western Wyoming. Here we are interested in testing whether metals accumulate differently in burned forests and unburned forests. We expect that metal concentrations will be highest in burned forests and open meadows due to higher rates of snow accumulation in the open canopy in winter months. We are also interested in the regional variability of metals in the study sites located around Jackson, Wyoming. We expect fire sites closest to Jackson to have the highest concentrations due to their proximity to a more urbanized landscape and thus, emission sources.

MATERIALS AND METHODS

2.1 Study Site

Samples were collected from a chronosequence of eight forests in the Triple Divide region of western Wyoming that extends around Jackson, Wyoming and includes the headwaters of the Columbia, Colorado, and Missouri rivers (Figure 1). The climate in Jackson is cold and temperate, with an average annual temperature of 3.6℃ and approximately 851 mm of precipitation (climate-data.org). The forests are pine-dominated, predominantly lodgepole pine (*Pinus contorta)* and whitebark pine (*Pinus albicaulis*). Historically, forest fires are frequent and regular across the seasonal snow zone. The fires used in this study occurred between 2000 to 2018 with mixed burn severity in forests of similar elevation, forest structure, composition, and topography (Figure 1).

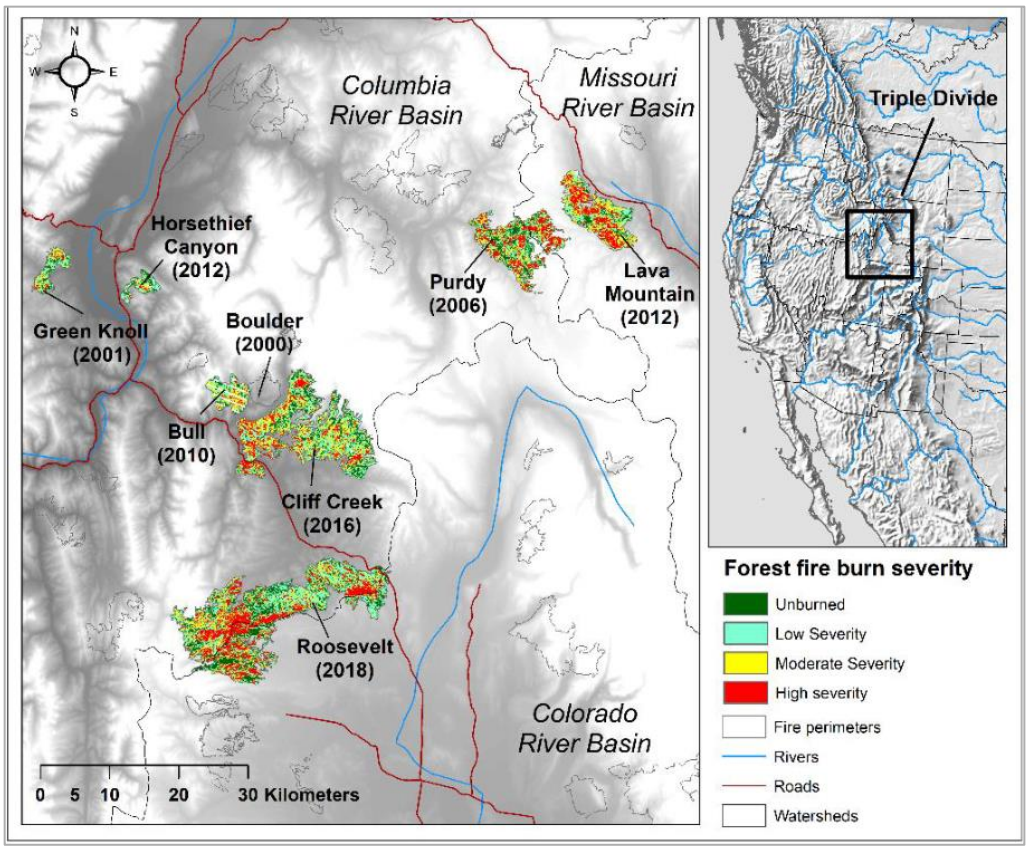

Figure 1: The study area is located in the Triple Divide region that includes the headwaters of the Columbia, Colorado, and Mississippi rivers (adapted from Gersh et al, 2022).[18]

Table 1: Characteristics of eight forest fires used in this study.

Fire Name	Ignition Date	Elevation (m)	Major River System	Average Burn Severity
Boulder - BOF	31 July 2000	2240	Columbia	Moderate
Green Knoll - GKF	22 July 2001	2144	Columba	Moderate
Purdy - PF	4 August 2006	2804	Columbia/Missouri	Moderate
Bull - BF	23 July 2010	2164	Columbia	Moderate
Horsethief Canyon - HTCF	8 September 2012	2286	Columbia	Low
Lava Mountain - LMF	11 July	2432	Missouri	Moderate
Cliff Creek - CCF	17 July 2016	2225	Columbia	Moderate
Roosevelt - ROF	15 September 2018	2438	Columbia/Colorado	Moderate

2.2 Geochemical Analysis and Statistics

We evaluated 397 snow core samples that were collected in February of 2019 and 2020 at eight sampling sites (Table 1). Snow samples were melted at room temperature for 24 hours or less, then preserved in 1% nitric acid (Optima Grade, Fisher Chemical) for 3 months or longer.[19] Due to the large sample size, trace element concentrations were determined in six separate batches using an Agilent Technologies 7900 inductively coupled

plasma mass spectrometer. ICPMS replicated measurements three times for each sample to produce a relative standard deviation (RSD) value for each sample. We performed calibration using multi-element standards (SPEX CertiPrep Multi-element Solution 4). All calibration standards were within 15% of the expected concentrations. Quality assurance was conducted every 10 to 15 samples (Inorganic Ventures IV-ICPMS-71A). A quality assurance summary is given in Supplemental Table 1. Replicate samples were also measured every 10 to 15 samples to monitor instrument drift. Detection limits varied depending on the element.

2.3 Data Analysis and Statistics

Non-detects complicate statistical analysis, and throwing out non-detected values or dropping them to zero can introduce bias when analyzing summary statistics. To address this, we reported non-detects as one-half the detection limit as per the Groundwater Statistics and Monitoring Compliance method[20]. We threw out measurements with high RSD (>20%) and measurements that were far outside calibration limits (> 250 µg/L). We log transformed non-normal data and performed a nested ANOVA and Tukey's HSD. Some element distributions were not normal or equal even after log transformation, so we used the non-parametric Kruskal-Wallis tests to detect differences in concentrations between fires and burn severities.

RESULTS

3.1 Regional results

The ICP-MS analysis revealed some differences in average metal concentrations between the eight forest fire sites sampled in Wyoming. Results for six significantly measured analytes are listed in Table 2. The Horsethief Canyon (HTCF) site stood out as having the highest mean concentrations of Al at 84.92 ± 65.88 µg/L, V at 0.17 ± 0.10 µg/L, Cr at 0.31 ± 0.22 µg/L, As at 0.09 ± 0.07 µg/L, and Pb at 0.19 ± 0.19 µg/L (p-values <0.05) (Figure 2). In contrast, the Bull Fire (BF) site exhibited significantly lower mean concentrations of the six elements (Figure 2). Manganese was not statistically different at the HTCF site, but it was significantly different at the Bull Fire site.

Table 2: Median, IQR, mean, and standard deviation (µg/L) of aluminum (Al), manganese (Mn), chromium (Cr), vanadium (V), arsenic (As), and lead (Pb) at each fire site (µg/L)

	Boulder	Green Knoll	Purdy	Bull	Horsethief Canyon	Cliff Creek	Lava Mt.	Roosevelt
(ug/L)								
Al								
Median	20.74	40.74	16.83	2.84	82.56	21.64	22.72	13.88
IQR	33.47	49.74	20.55	7.29	91.81	27.06	14.51	19.48
Mean	5.97	50.20	24.58	5.97	84.92	33.98	24.02	18.90
SD	6.99	45.36	19.33	6.70	65.88	32.81	0.03	22.92
V								
Median	0.06	0.11	0.05	0.03	0.18	0.07	0.06	0.05
IQR	0.06	0.10	0.03	0.02	0.17	0.06	0.01	0.02
Mean	0.08	0.14	0.06	0.03	0.17	0.09	0.06	0.05
SD	0.07	0.22	0.03	0.02	0.10	0.06	0.01	0.03
Cr								
Median	0.12	0.12	0.09	0.05	0.24	0.09	0.14	0.06
IQR	0.18	0.14	0.08	0.07	0.25	0.12	0.11	0.08
Mean	0.15	0.18	0.18	0.07	0.31	0.15	0.28	0.07
SD	0.16	0.29	0.18	0.09	0.22	0.11	0.35	0.09
Mn								
Median	1.54	2.27	1.12	0.37	4.89	1.74	1.86	2.85
IQR	1.73	1.97	1.13	0.80	4.02	1.50	1.50	2.88
Mean	4.50	5.49	2.02	0.61	5.07	2.72	2.90	6.11
SD	13.95	21.56	3.74	0.88	2.67	3.57	4.79	16.17
As								
Median	0.02	0.04	0.01	0.00	0.07	0.02	0.02	0.01
IQR	0.04	0.06	0.40	0.01	0.09	0.06	0.01	0.04
Mean	0.04	0.07	0.03	0.02	0.09	0.04	0.03	0.03
SD	0.06	0.14	0.04	0.04	0.07	0.05	0.04	0.05
Pb								
Median	0.06	0.12	0.03	0.01	0.16	0.02	0.05	0.02
IQR	0.13	0.27	0.06	0.01	0.20	0.12	0.07	0.02
Mean	0.01	0.19	0.07	0.03	0.19	0.08	0.11	0.04
SD	0.12	0.24	0.13	0.08	0.19	0.10	0.24	0.06

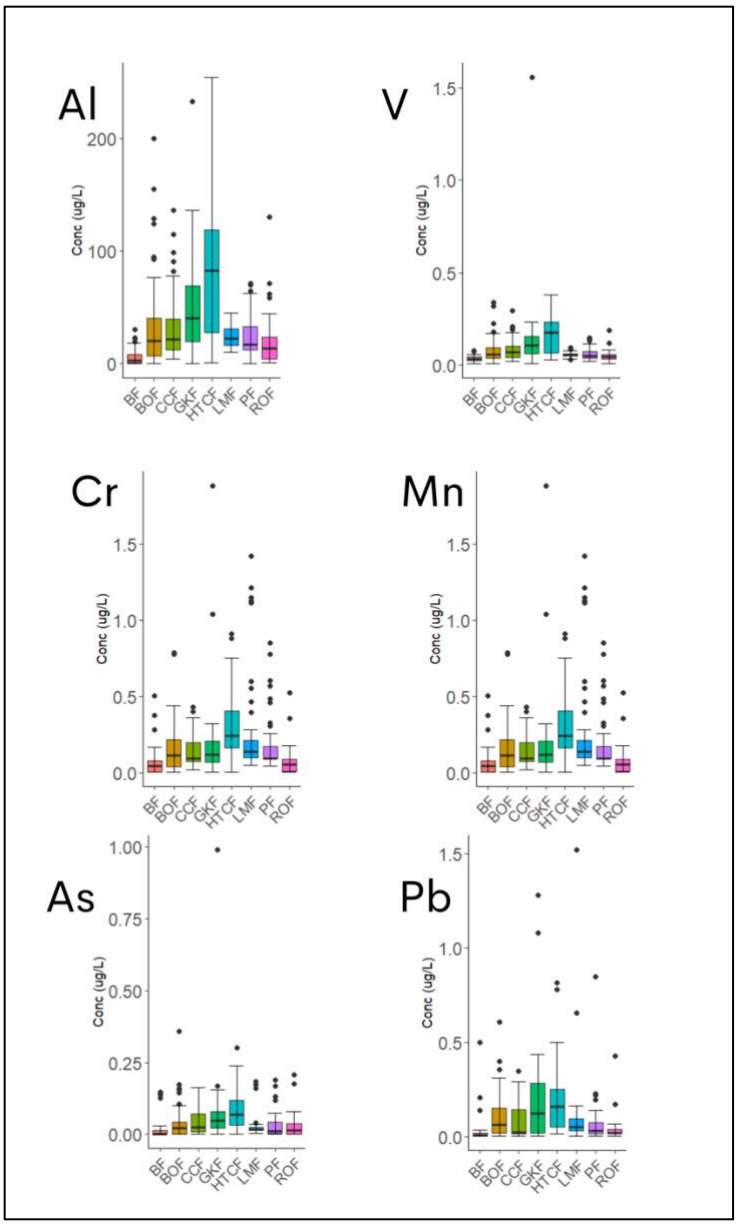

Figure 2: Boxplots of aluminum (Al), vanadium(V), chromium (Cr), manganese (Mn), arsenic (As), and lead (Pb) that were detected (µg/L) that were significantly higher in Horsetheif Canyon (HTCF)(p-values <0.05) and lower in Bull Fire (BF) (p-values < 0.05).

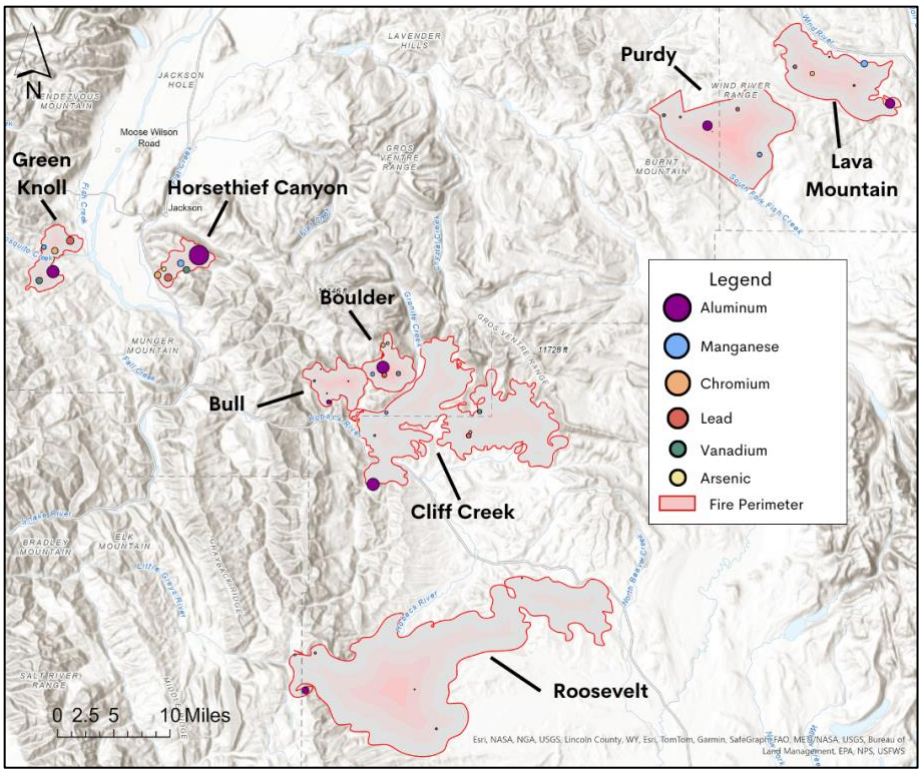

Figure 3: Map of average concentrations (ug/L) of Al, Mn, Cr, Pb, V, and As in each fire site. Larger dots indicate higher average concentrations. Points were randomly assigned to the fire boundary and do reflect specific locations within the fire site.

3.2 Forest structure results

Six elements showed significant differences among the three groups. Those elements, in order of highest to lowest concentration, were Al> Mn> Cr > Pb > V > As. High variability and very low concentrations for elements Zn, Ni, Cu, Mo, Se, and Cd limited statistical signals, and we found no significant differences between burned and unburned forests for these elements. In general, unburned forests had the highest levels of six elements, open areas had the lowest concentrations, and burned forests exhibited intermediate values. We observed high variability within each forest structure category. In some cases, the standard deviation exceeded the mean value, suggesting a non-normal distribution, which was accounted for by using the non-parametric Kruskal-Wallis test to detect differences among groups. We reported median, interquartile range, mean, and standard deviations for the six detected elements in Table 2.

In general, forest concentrations were significantly higher than unburned forests for the six elements (p-values <0.05). We visualized the differences on the log scale to show the spread of data better and reduce the effects of extreme outliers (Figure 4). Aluminum levels were nearly double in unburned forests at 57.35 ± 46.5 µg/L (p-value < 0.001) than burned forests at 27.5 ± 39.3 µg/L and open areas at 24.2 ± 29.8 µg/L (Table 3). Manganese showed a similar pattern, but at lower concentrations with the highest mean value of 7.06 ± 16.80 µg/L in unburned forests compared to 3.26 ± 11.31 µg/L in burned forests and 1.84 ± 2.47 µg/L in open areas respectively

(p-value < 0.001). Lead concentrations averaged 0.15 ± 0.08 µg/L in unburned forests, nearly double the levels found in burned forests (0.09 ± 0.18 µg/L) and open areas (0.08 ± 0.12 µg/L). Arsenic showed a comparable trend, with average values of 0.07 ± 0.06 µg/L in unburned forests, 0.04 ± 0.09 µg/L in burned forests, and 0.03 ± 0.04 µg/L in the open sites. Average vanadium concentrations were 0.12 ± 0.08 µg/L in unburned forests, 0.08 ± 0.12 µg/L in burned forests, and 0.06 ± 0.05 µg/L in open areas. Chromium levels were more similar across forest types, but still followed a statistically significant trend (p-value < 0.05) of unburned forests being higher than burned and open forests. Average chromium in unburned forests was 0.18 ± 0.16 µg/L, 0.17 ± 0.25 in burned forests, and 0.16 ± 0.19 µg/L in open areas. Burned forests and open areas across all six elements were not statistically different (p-value > 0.05). Overall, results from statistical analysis indicate that for six elements, there is a consistent pattern of higher metal accumulation in unburned forests compared to burned forests and open areas, though concentrations were variable (Figure 2)

Table 3: Significantly higher concentrations of six trace elements (µg/L) in all snow core samples in unburned forests (n=8) compared to burned forests (n=8) and open meadows (n=8) in western Wyoming.

	Burned	**Forest**	**Open**
Al (µg/L)			
Median	15.52	41.71	15.69
IQR	23.55	67.27	22.08
Mean	27.52	57.35	24.40
SD	39.32	46.5	29.83
V (µg/L)			
Median	0.05	0.09	0.05
IQR	0.04	0.12	0.03
Mean	0.08	0.12	0.06
SD	0.12	0.08	0.05
Cr (µg/L)			
Median	0.09	0.14	0.09
IQR	0.11	0.16	0.12
Mean	0.17	0.18	0.16
SD	0.25	0.16	0.19
Mn (µg/L)			
Median	1.57	3.48	1.15
IQR	2.08	3.59	1.71
Mean	3.26	7.06	1.84
SD	11.31	16.80	2.47
As (µg/L)			
Median	0.02	0.04	0.02
IQR	0.03	0.07	0.05
Mean	0.04	0.07	0.03
SD	0.09	0.06	0.04
Pb (µg/L)			
Median	0.03	0.09	0.03
IQR	0.09	0.22	0.07
Mean	0.09	0.15	0.08
SD	0.18	0.08	0.12

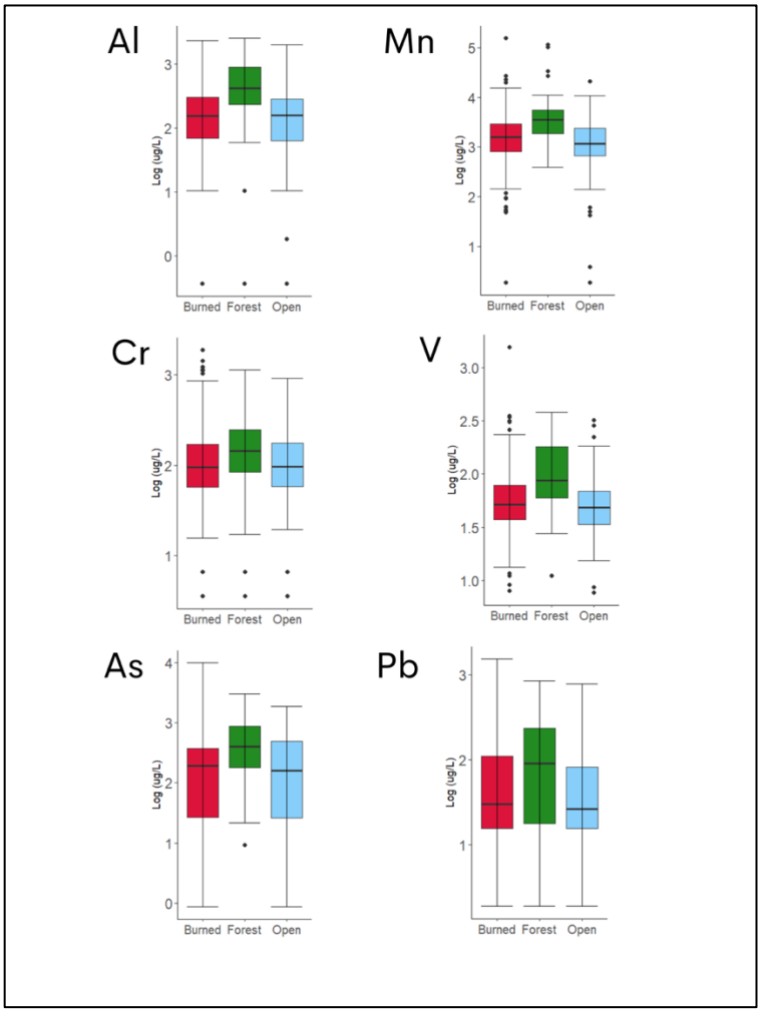

Figure 4: Logarithmic-scale median concentrations ± min and max and IQR. Significant differences exist between unburned forests than burned forests, and unburned forests and open meadows (p- value < 0.05) in eight fire sites in western Wyoming. No significant differences exist between burned forests and open meadows (p-value > 0.05).

DISCUSSION

4.1 Long-range atmospheric transport in remote areas

The results of this study provide insight into the variation of trace element concentrations in snowpack across a chronosequence of forest fires sites in a remote area. Horsethief Canyon, the fire site just south of Jackson, WY, exhibited the highest number of elevated concentrations for five elements in the following order: Al > Cr > Pb > V > As. The unique chemical signatures and observed spatial north to south gradient of concentrations could be attributed to long-range atmospheric transport from local or distant emission sources, which is consistent with findings from Bing et al. [21,22] who reported that atmospheric transport is a major source of trace metal contamination in remote mountainous areas. Horsethief Canyon is an exposed west-facing slope

just south of Jackson, WY, and is subject to prevailing westerly winds. The topographic features of the canyon may act as an orographic barrier, potentially "collecting" elements from distant emission sources.

The high variability observed in each site suggest that factors other than the fire site's proximity to emission sources could influence trace element concentrations in the snowpack. Bing et al. suggest that the distribution and variability of metals can be influenced by altitude, plant interception, vegetation composition, and other more localized meteorological or topographical factors.[15] Future work should aim to investigate these factors in more detail to better understand the processes driving the spatial and temporal distribution of metals in remote areas of western Wyoming.

4.2 Canopy slows wind velocity

The observed differences in average concentrations of Al, Mn, Cr, Pb, V, and As between burned forests, unburned forests, and open areas suggest that forest structure plays a significant role in the deposition and accumulation of metallic elements in forested watersheds. Higher concentrations observed in unburned forests could be attributed to several factors. First, research by Oliver[23] indicates that wind velocity slows over a forest canopy due to the increased surface roughness and drag exerted by the vegetation below.[16] The slower moving air may allow for more time for the particles to deposit onto the forest floor or snowpack surface. Additionally, Moon et al.[24] found that regrowth open forests with reduced canopies experience faster wind velocities and turbulence than dense forests. Higher velocity winds have a higher carrying capacity for particles.[25] Because wildfires remove forest canopy, we assume that they act more like open canopy forests with respect to wind due to reductions in vegetation surface roughness. As the faster moving air moves through the open canopy, the particles may be carried elsewhere until they meet some obstacle that allows the air to slow down and deposit snow.

4.3 Potential speciation in throughfall

While this study did not specifically account for throughfall chemistry, He et al. [26] discuss that the presence of forest canopy can intercept and retain atmospheric particles over time, leading to higher accumulation of metals in forested areas compared to open areas. Thus, the observed increase in metal concentrations in unburned forests may be related to the longer exposure time of forests to atmospheric deposition compared to burned forests. Some elements are speciated in the throughfall process. Gandois et al.[27] found that concentrations of certain metals in rain below a forest canopy was significantly higher than the precipitation above the canopy, indicating as precipitation moves through a forest canopy, it becomes more concentrated depending on the element. Lastly, Bing et al. [22] reported that coniferous forests have a higher retention capacity for metals than deciduous forests. The Triple Divide region is dominated by

pine species, which could play a role in the accumulation and retainment of metals within the forest canopy and subsequent snowpack. Further, our findings preliminarily support that the reduction in forest canopy may lead to decreases in metal accumulation in winter snowpacks. The burning of a forest canopy may have liberated accumulated metals back into the atmosphere to be deposited elsewhere. Specifically, releases of Al and Mn in ash have been shown to have a strong relationship with burn severity, while release of Zn is more related to variations in topography.[28]

4.4 Forests collect pollutants

Extensive research has demonstrated the effectiveness of forests in cleaning the atmosphere, which is an important ecosystem service.[29] Our findings align with this research, suggesting that forests excel at accumulating atmospheric pollutants. While this is beneficial from an atmospheric pollution perspective, it may have negative consequences for the hydrosphere and the health of watersheds oncet the snow melts. High concentrations of pollutants in forests, especially forests close to urban centers, could lead to a pulse of contaminants flowing through forested watersheds and into aquatic systems. To better understand the movement of pollutants between the atmosphere and hydrosphere and lithosphere, an analysis of spring-time snowmelt and water quality downstream would be valuable.

CONCLUSION

Our results indicate a need for more a comprehensive understanding of the numerous factors that influence the deposition and accumulation of trace elements in snow following forest fire events. Because wildfires are increasing in extent, duration, and severity across the western U.S., our findings reveal the potential impact of wildfires on the cycling of metals in remote forest ecosystems. The similarities between metal concentrations in burned forests and open meadows suggests that the removal of forest canopy by wildfire may change spatial nature of atmospheric deposition. Our findings also reveal that while forests clean the air, they may also be concentrating metal pollution in snowpack that eventually ends up in the surrounding watershed and aquatic systems. Our findings can potentially help inform future forest management decisions related to monitoring of hydro-chemical cycles relative to forest disturbance in a rapidly changing climate.

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SUPPLEMENTARY TABLES

Supplemental Table 1: Quality control recovery (%) for 25ppb for six ICP-MS runs.

	Al	V	Cr	Mn	Ni	Cu	Zn	As	Se	Mo	Cd	Pb
QC-1												
	104	94	95	97	96	111	108	93	94	93	94	100
	103	95	96	97	96	111	107	93	93	93	94	98
	103	95	96	97	96	111	109	94	95	93	94	99
	110	99	100	102	98	115	116	97	98	95	96	100
QC-2												
	111	99	101	100	100	105	103	98	98	98	101	101
	120	103	107	107	105	112	112	103	104	100	102	100
	117	102	105	105	103	109	109	101	102	98	101	98
	121	106	108	108	106	114	115	104	106	100	103	98
QC-3												
	108	99	98	102	101	105	103	99	98	97	98	104
	103	96	94	99	97	100	96	95	92	92	93	97
	100	93	93	96	94	96	92	92	89	90	91	94
	100	93	91	96	93	96	91	92	89	89	90	94
QC-4												
	110	107	108	108	108	112	107	107	107	108	110	112
	108	103	105	105	104	107	103	103	102	99	103	103
	104	99	101	100	99	103	99	99	98	95	98	97
	94	93	94	95	92	97	91	92	92	88	90	89
QC-5												
	95	93	93	94	95	94	NA	94	92	91	93	92
	95	93	93	94	95	94	NA	94	92	91	93	92
	98	94	94	95	93	92	NA	93	92	90	91	89
	97	93	93	92	92	91	NA	92	90	88	90	87
QC-6												
	122	105	106	108	107	108	96	105	104	106	111	109
	117	98	100	102	101	104	89	100	98	97	100	97
	112	98	98	100	98	100	88	98	97	95	99	94
	109	95	98	101	98	99	86	96	96	94	98	94