River sediment geochemistry and provenance following the Mount Polley mine tailings spill, Canada: the role of hydraulic sorting and sediment dilution processes in contaminant dispersal and remediation.

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ABSTRACT

The failure of the Mount Polley tailings storage facility (TSF) in August 2014 was one of the largest magnitude failures on record, and released approximately 25 Mm^3 of material, including c. 7.3 Mm^3 of tailings into Hazeltine Creek, part of the Quesnel River watershed. This study evaluates the impact of the spill on the geochemistry of river channel and floodplain sediments and utilizes Pb isotope ratios and a multi-variate mixing model to establish sediment provenance. In comparison to sediment quality guidelines and background concentrations, Cu and V were found to be most elevated. Copper in river channel sediments ranged from 88-800 mg kg^{-1}, with concentrations in sand-rich and clay/silt-rich sediments being statistically significantly different. Concentrations in river channel were believed to be influenced by hydraulic sorting during the rising and falling limbs of the flood wave caused by the tailings spill. Results highlight the importance of erosive processes, instigated by the failure, in incorporating soils and sediments into the sediment load transported and deposited within Hazeltine Creek. In this instance, these processes diluted tailings with relatively clean material that reduced metal concentrations away from the TSF failure. This does however, highlight environmental risks in similar catchments downstream of TSFs that contain metal-rich sediment within river channels and floodplain that have been contaminated by historical mining.

KEY WORDS: tailings, spill, metals, lead isotopes, fingerprint

HIGHLIGHTS

- Copper concentrations exceed sediment quality guideline level following the spill.
- Hydraulic sorting influenced spatial trends in metal concentrations.
- Lead isotopes used to fingerprint sediments after the tailings spill.
- Mixing model data indicate the importance of spill-induced erosive processes

INTRODUCTION

Mine tailings are the milled solid waste left over from the recovery of the valuable commodities from mined material (Kossoff et al., 2014). Although the chemical properties of mine tailings can vary substantially, the material represents the most voluminous metalliferous waste produced by metal mines (Lottermoser, 2010). However, the volume of tailings compared to unmilled waste rock, produced during mining, may be lower and will vary between surface and underground mines. Despite the growth of other storage approaches, currently the majority of mine tailings are transported and stored as a slurry, with tailings storage facilities (TSFs) representing substantial pieces of mine site infrastructure. Worldwide, there are estimated to be over 12,000 TSFs (Macklin et al., submitted), of varying construction type and in numerous mining operations the land area covered by TSFs now exceeds that being used for mining activities (Hudson-Edwards et al., 2011).

Since 1960 there have been a reported 158 mine tailings dam failures worldwide (Project), 2020). It is therefore apparent that these structures represent a substantial global risk to the environment and local populations. The environmental impacts of the failure of tailings dams, associated with the release of large volumes of metal-rich tailings and water into recipient environments, have been noted by many studies over the last two decades (Hudson-Edwards, 2016; Hudson-Edwards et al., 2003; Macklin et al., 2003). Of concern is the potential that the frequency of such events may increase over the coming years, due to a) the growing number of active and inactive tailings ponds, driven by higher waste to ore ratios, as high-grade ores are exhausted (Mason et al., 2011), and b) an increase in extreme hydro-meteorological events, a common contributor to many failures (Rico et al., 2008).

The tailings:water ratio commonly varies among failure events (Rico et al., 2008), and the volume of tailings released can have an important influence on 1) approaches to post-event remediation, 2) the geomorphological disturbance within the recipient river systems, and 3) the longer-term fate of metals released into recipient environments. Furthermore, the chemical nature of spilled material varies considerably (Kossoff et al., 2014), reflecting the mineralogy of the ore-body from which the tailings derive, the efficiency of the extraction process and any substances used in ore processing (for example CN^- in the case of Au extraction). However, what is common is that mine tailings dam failures represent a major environmental risk with the potential to impact river systems in terms of geomorphology, geochemistry and ecosystem health (Kossoff et al., 2014; Macklin et al., 2006).

The partial embankment breach at the Mount Polley TSF on 4th August 2014, is the second largest mine waste spill by volume on record (Project), 2020). The causes of the spill have been reported in detail elsewhere (Byrne et al., 2018; Hudson-Edwards et al., 2019). The spill resulted in approximately 25 Mm^3 of material being released into the Quesnel River watershed (Petticrew et al., 2015). This comprised approximately 7.3 Mm^3 of tailings, 17.1 Mm^3 of supernatant and interstitial water and 0.6 Mm^3 of TSF materials (Petticrew et al., 2015). The release of water and sediment from the TSF created a flood wave that eroded the existing river valley and resulted in the deposition of material along the valley floor of Hazeltine Creek. Deposits were up to 3.5 m thick and extended up to 100 m from the river channel.

The impacts of the Mount Polley spill have been studied with respect to the influence on water quality in Hazeltine Creek (Byrne et al., 2018) and Quesnel Lake (Petticrew et al., 2015) and the release of Cu and V, specifically, into the environment (Hudson-Edwards et al., 2019). In addition, data have been published on the geochemistry of the mine tailings (Kennedy et al., 2016). To date, however, there has been no published study into the fate of particulate material released by the spill, and in particular the mixing and subsequent deposition of released tailings and eroded valley floor sediments. These factors are crucial to understanding the storage of tailings and longer-term environmental impacts of the spill. To this end, this study aims to utilize geochemical fingerprinting to understand the contribution of different source materials within the Hazeltine Creek catchment and to quantify their influence on post-spill river sediment dynamics. Our primary objective was to quantify the contributions of different types of tailings material released by the spill and to establish which contributed most significantly to river sediment contamination. Our expectation was that a better understanding of the fate of material released by the spill and by erosive processes generated by the post-spill flood, will help to provide a better understanding of the potential legacy of tailings dam failures.

STUDY AREA

The Mount Polley deposit is an alkalic porphyry Cu-Au deposit (Panteleyev, 1995), formed approximately 180 Ma ago within Late Triassic (235 – 201 Ma) and Mesozoic (252 – 66 Ma) bedrock geology (Kennedy and Day, 2015). Sulfide mineralization consists principally of chalcopyrite ($CuFeS_2$) and pyrite (FeS_2) but, at least 50% of the Cu mineralization is not sulfidic, and includes primarily malachite ($Cu_2CO_3(OH)_2$) and chrysocolla ($(Cu,Al)_2H_2Si_2O_5(OH)_4.nH_2O$) (Henry, 2009). Overall, the tailings produced at Mount Polley have a low sulfide content (0.1-0.3 wt. %) and are not acid-generating (Kennedy and Day, 2015), making the Mount Polley event unusual compared to many other tailings spills (WISE, 2020).

The deposit is located approximately 55 km north-east of Williams Lake, British Columbia, within the 112 km^2 Hazeltine Creek catchment (Figure 1). Hazeltine Creek is a tributary catchment within the larger Quesnel River Catchment, and flows for 10.3 km from the southern end of Polley Lake at 920 m asl, into Quesnel Lake at 730 m asl (Burge and Cuervo, 2015).

Mining began at Mount Polley Mine in 1997 and 95 M tonnes of ore were processed between the commencement of mining and 2014, producing 94.2 M tonnes of tailings in the same period (Kennedy and Day, 2015). Concentrations of most metals within the tailings are relatively low, but the material is relatively enriched in Cu and V (86-296 and 8-55 mg kg^{-1}, respectively) (Kennedy and Day, 2015).

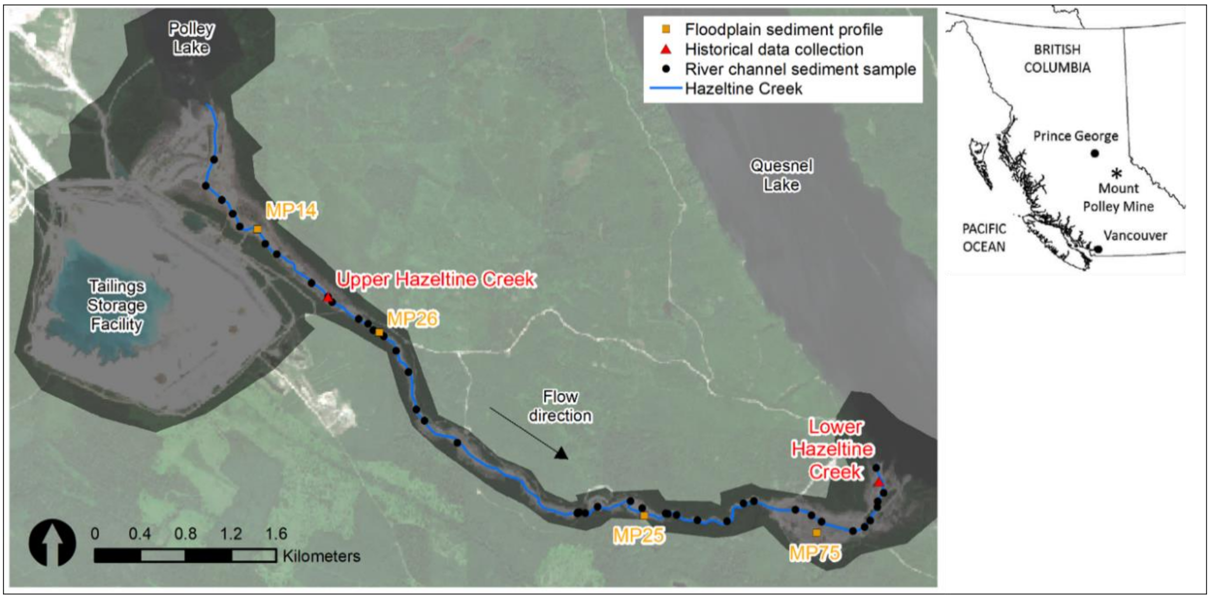

Figure 1. Map showing the study area including the location the Mount Polley TSF and sample sites used for the collection of river channel sediment and floodplain sediment and the location of longer-term sampling in Hazeltine Creek.

MATERIALS AND METHODS

Extensive geochemical datasets were provided by the Mount Polley Mining Corporation based upon the analysis of samples of tailings and stream sediments collected following the tailings spill (Minnow Environmental Inc, 2015; SRK Consulting, 2015). These data were utilized in addition to data produced by this study from the analysis of samples: 1) collected by this study and 2) samples collected for, and provided to the authors by the Mount Polley Mining Corporation (Mount Polley Mining Corporation, 2015).

In July and August 2015 determinations of metal concentrations in river channel and floodplain sediment from Hazeltine Creek and mine tailings deposited in the Hazeltine Creek river corridor were made in the field at 86 sites, using a portable x-ray fluorescence (pXRF) (Niton XLp 300) with an analysis time of 60 seconds. At 46 of these sites, samples of river channel and floodplain sediment from Hazeltine Creek, and deposited tailings, were collected for subsequent laboratory analysis of metal concentrations and Pb isotopes (Figure 1). Sampling of floodplain material was carried out at varying depths from exposed river bank profiles at sites MP14, MP25, MP26 and MP75 (Figure 1). In all instances, samples were collected as composite samples using a stainless steel trowel. Composite samples comprised 5-10 sub-samples collected over a c. 1 m^2 area (river channel sediments) or from the same floodplain depth, bulked together to form a single sample of c. 500 g.

In the laboratory, all samples were air-dried at 30 °C, disaggregated using a pestle and mortar and sieved to isolate the < 2mm fraction. Samples were digested in concentrated aqua regia (HCl and HNO_3 in a 3:1 v/v ratio) prior to multi-elemental analysis by Inductively Coupled Plasma – Mass Spectrometry (ICP-MS). The accuracy and precision of multi-element analyses was monitored through the analysis of a certified reference material (GSD-6, a stream sediment near an area of porphyry Cu mineralization), and the resultant data are presented in Supplementary Material 1. The aqua regia digestion matches the method was used in this study to provide consistency with methods used to generate the datasets provided by the Mount Polley Mining Corporation. To monitor the comparability of datasets provided by the Mount Polley Mining Corporation and those generated by this study, 20 randomly selected samples previously analysed by the Mount Polley Mining Corporation, were reanalysed by this study using the methods outlined above. The mean differences in concentrations between the duplicate analyses ranged from 9.3-27.7% (Supplementary Material 1) with greater variability generally found in samples with low element concentrations.

Lead isotopes ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb in a selection of sediment samples were determined by Magnetic Sector ICP-MS (Thermo-Finnegan Element2) at Aberystwyth University. Solutions for analysis were prepared at 50 ng ml^{-1} and analysed in batches along with blank samples (2 per batch) and NIST 981 reference material (2 per batch). Analytical precision was found to be 0.12 % (^{206}Pb/^{207}Pb), 0.18 % (^{208}Pb/^{206}Pb), 0.25 (^{208}Pb/^{204}Pb), 0.15 (^{207}Pb/^{204}Pb) and 0.16 (^{206}Pb/^{204}Pb).

Analytical accuracy versus the NIST 981 reference material was found to be 0.19 % (^{206}Pb/^{207}Pb), 0.09 % (^{208}Pb/^{206}Pb), 0.27 (^{208}Pb/^{204}Pb), 0.17 (^{207}Pb/^{204}Pb) and 0.26 (^{206}Pb/^{204}Pb).

A mixing model was used in order to quantify the contributions of mining and non-mining sources to the river channel and floodplain sediments present in Hazeltine Creek after the spill. The principles of the mixing model approach have been described in detail by Yu and Oldfield (1989) and Collins et al. (1997), and the range of model approaches that have been developed, and the geochemical properties of sediments used within the models, have been reviewed by Haddadchi et al. (2013). Given the small spatial scale of the study catchment and limited number of potential sources within it, this study utilized a model based upon Pb isotope signatures as fingerprint properties (Miller et al., 2007). In short, the approach utilises the following equation:

$$b_j = \sum \frac{m}{i} = 1 X_i \, a_{ij} \qquad \text{(Equation 1)}$$

where b_j ($_{j}$=1,2,3,4) are Pb isotope ratios (^{206}Pb/^{207}Pb, ^{208}Pb/^{206}Pb, ^{208}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, ^{206}Pb/^{204}Pb) of a stream sediment sample composed of *m* distinct source materials (Table 1), a_{ij} ($_{i}$=1,2,3…,m) are the corresponding Pb isotope ratios of the *i*th source materials and X_i being the proportion if the *i*th source in the sediment. Given values of b_j and a_{ij}, a series of *n* linear equations were optimized using the Solver function in Microsoft Excel to quantify the contributions of the five sources identified. Two important constraints are that all source proportions must be non-negative (Equation 2) and source proportions must sum to unity (Equation 3).

$$X_i \ = \geq \ 0 \qquad \text{(Equation 2)}$$

$$\sum_i^m = \ 1X_i = 1 \ \text{(Equation 3)}$$

The validity of the mixing model results was assessed by comparing the measured parameter values in the sediment with the values predicted in the optimization of the linear equations (Equation 4). This assessment quantifies relative errors and indicates whether the mixing model generates an acceptable prediction of the fingerprinting properties (Miller et al., 2007). Errors for five iterations of the mixing model (one per source group) ranged from 0.27-0.54%.

$$\% \, error = \sqrt{\frac{\left(\sum_{i=1}^{m}\left(bi - \sum_{j=1}^{n} a_{ij}x_j\right)\right)^2}{\sum_{i=1}^{m}(b_i)^2}} \times 100 \qquad \text{(Equation 4)}$$

Potential sources of uncertainty within mixing model approach have been summarized and discussed by Collins et al. (2010; 2012). These include the potential for statistically similar solutions during the optimization process, especially close to 0 and 100% sediment contribution, and the possible variability in fingerprint properties that is not captured by the analysis of samples of the source materials. The relative errors produced by the mixing model are small, although mindful of these potential uncertainties, the mixing model should be seen as providing a general insight to sediment contributions and therefore interpreted in terms of broader trends.

Table 1. Source materials (source group) used to establish river sediment provenance.

Source material / source group	Description
Background sediments (as termed by Mount Polley Mining Corporation). Analysis of 12 samples.	Sediments from the Hazeltine Creek valley floor beyond the extent of material deposited by the spill. Samples provided by Mount Polley Mining Corporation and analysed by this study.
Native channel sediments (as termed by Mount Polley Mining Corporation). Analysis of 3 samples.	Material from the Hazeltine Creek channel banks that was not covered by, or disturbed by the spill. Reflects material present in Hazeltine Creek prior to the spill event. Samples provided by Mount Polley Mining Corporation and analysed by this study.
Sand-rich tailings. Analysis of 3 samples.	Sand-rich tailings released by the spill. Samples provided by Mount Polley Mining Corporation and analysed by this study. Average sediment composition of analysed samples[a]: <0.1% gravel; 70% sand; 25.5% silt; 4.8% clay
Silt and clay-rich tailings. Analysis of 4 samples.	Fine grained, silt and clay-rich tailings released by the spill. Samples provided by Mount Polley Mining Corporation and analysed by this study. Average sediment composition of analysed samples[a]: <0.1% gravel; 16.1% sand; 67% silt; 17% clay
Mixed tailings. Analysis of 2 samples.	A mixture of sand-rich and silt and clay-rich tailings released by the spill. Samples provided by Mount Polley Mining Corporation and analysed by this study. Average sediment composition of analysed samples[a]:

	<0.1% gravel; 52.1% sand; 36.8% silt; 11% clay

[a]Sedimentological data from previous analysis of the same sample material by Minnow Environmental Inc (2015) and SRK Consulting (2015).

Finally, selection of river channel sediment (n = 8) and floodplain sediment samples (n = 6), specifically, those used for the mixing model, were also analysed for their sedimentological composition. The % proposition of sand (2 mm – 63 µm), silt (63 – 3.9 µm) and clay (< 3.9 µm) was determined, firstly by sieving to separate the sand and combined silt/clay fraction, and secondly using a Mastersizer 2000 to determine the relative proportions of silt and clay following Malvern Instrument's standard protocol (Malvern Instruments Ltd, 2007). Gravel-sized material (> 2mm) was not present in any samples analysed.

RESULTS AND DISCUSSION

Metals in mine tailings

Tailings released by the spill comprised two types of material: first a sand-rich material ('sandy tailings', ST), comprising an average 73% sand, 26% silt and clay and 1% gravel (Minnow Environmental Inc, 2015; SRK Consulting, 2015); and second finer-grained material ('fine-grained' tailings, FT), comprising an average 38% sand, 61% silt and clay and 1% gravel (Minnow Environmental Inc, 2015; SRK Consulting, 2015). The two material types represent different products of ore processing.

Copper and V were generally the most enriched trace metals within both types of tailings (Figure 2), reflecting the nature of mineralization at Mount Polley, which is also reflected in the regional and local background geochemistry (Table 2). Average Cu and V concentrations were higher in the sandy tailings (1000 mg kg^{-1} and 195 mg kg^{-1}, respectively) compared to the finer-grained tailings (808 mg kg^{-1} and 170 mg kg^{-1}, respectively). In comparison to Cu, V concentrations exhibited less variability in both tailings materials. Average concentrations in the tailings of other potentially harmful elements, such as As, Cd, Pb and Zn (Figure 2) were also above average background concentrations determined for the British Columbia region and for the Hazeltine Creek catchment.

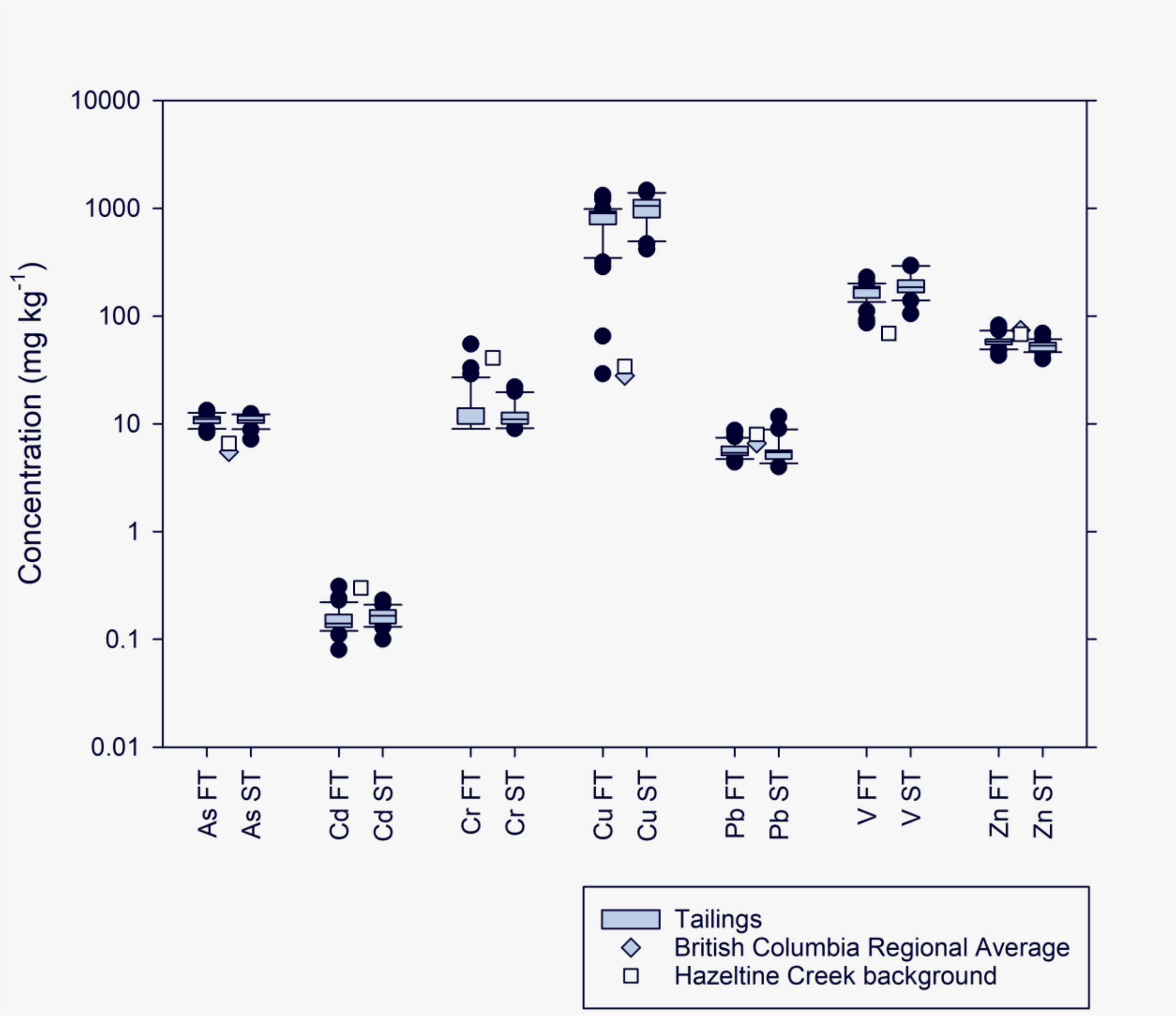

Figure 2. Summary of metal and As concentrations in clay and silt-rich tailings (‘fine-grained’ tailings [FT]) and sand-rich tailings (‘sandy’ tails [ST]). Number of samples: n = 41 (fine-grained tailings) and n = 20 (sandy tailings). Data adapted from SRK Consulting (Canada) Inc. (2015). Data also plotted for British Columbia regional average (As, Cu, Pb Zn only) (Geological Survey of Canada, 1981) and background concentrations (Minnow Environmental Inc., 2015).

Table 2. Minimum, mean, median and maximum background concentrations (mg kg^{-1}) determined for the Hazeltine Creek catchment and the British Columbia region (As, Cd, Cu, Pb and Zn only).

	Arsenic	Cadmium	Copper	Lead	Vanadium	Zinc
		Hazeltine Creek background[a]				
Minimum	3	<0.1	6	4	40	32
Mean	7	0.3	34	8	69	68
Median	6	0.2	22	6	61	53
Maximum	14	2	135	22	133	149
		British Columbia regional background[b]				
Minimum	1	ND	0	1	ND	4
Mean	6	ND	29	7	ND	75
Median	3	ND	24	4	ND	54
Maximum	96	ND	701	96	ND	3701

	Hazeltine Creek native channel sediment[c]					
Minimum	0.4	<0.1	6.3	4.1	2	2.3
Mean	7	0.2	36	7.3	60	54
Median	7	0.2	32	6.4	61	51
Maximum	14.7	0.4	87	14	100	94

[a] Data for 26 soil samples from Hazeltine Creek catchment; adapted from Minnow Environmental Inc. (2015). Concentrations determined following aqua regia digest.

[b] Data for 1290 stream sediment samples adapted from the Geological Survey of Canada (1981). Concentrations determined following aqua regia digest.

[c] Data for 17 native channel sediment samples within Hazeltine Creek adapted from SNC-Lavalin (2015). Sampled from channel banks and was not covered by, or undisturbed by the spill, reflecting material present in Hazeltine Creek prior to the spill event. Concentrations determined following aqua regia digest.

ND = no data

Metals in river channel sediments

Concentrations of As, Cu, Pb and Zn in Hazeltine Creek river channel sediments are plotted for silt-rich and sand-rich sediments in Figure 3; all Cd concentration were non-detectable and are not plotted. Non-parametric significant difference analysis (Mann-Whitney U test) indicates that, although there is no significant difference between As (p=0.288), Pb (p=0.276) and Zn (p=0.283) concentrations in sand- and silt-rich samples, Cu concentration are significantly different (p=<0.000). The sedimentological analysis of a selection of river channel sediments indicated that silt-rich sediments (n = 4) were found to contain: 35-43 % sand, 56-63 % silt and 1-5 % clay. Sand-rich sediments (n = 4) were found to contain: 75-95 % sand, 4-11 % silt and 0.3-0.5 % clay (Figure 4a). Data indicate that channel sediments present within Hazeltine Creek are present in accumulations that have notably higher sand (sand-rich) or silt (silt-rich) contents (Figure 4a). The spatial trends suggest that sand- or silt-rich sediments have accumulated throughout the study reach, likely reflecting spatial variation in processes influencing sediment transport and deposition.

Concentrations are compared to British Columbia (BC) Sediment Quality Guidelines (SQG) (Table 3), which comprise a lower Threshold Effect Level (TEL) concentration and an upper Probable Effect Level (PEL) concentration and are based on those produced by the Canadian Council of Ministers of the Environment (CCME) (2017). In addition, background concentrations determined for Hazeltine Creek (Table 2) are also plotted for comparison. All As concentrations fall below the BC SQG PEL, whereas all Pb and Zn concentrations are below the lower BC SQG TEL. In contrast, Cu concentrations in 92% of samples were above the upper BC SQG PEL. It is important to note that SQGs do not consider the potential bioavailability of sediment-associated metals, for example, as influenced by their physico-chemical speciation. Therefore, concentrations above a guidelines value may pose a lesser or greater significant environmental risk depending on the degree of bioavailbility (Guan et al., 2018).

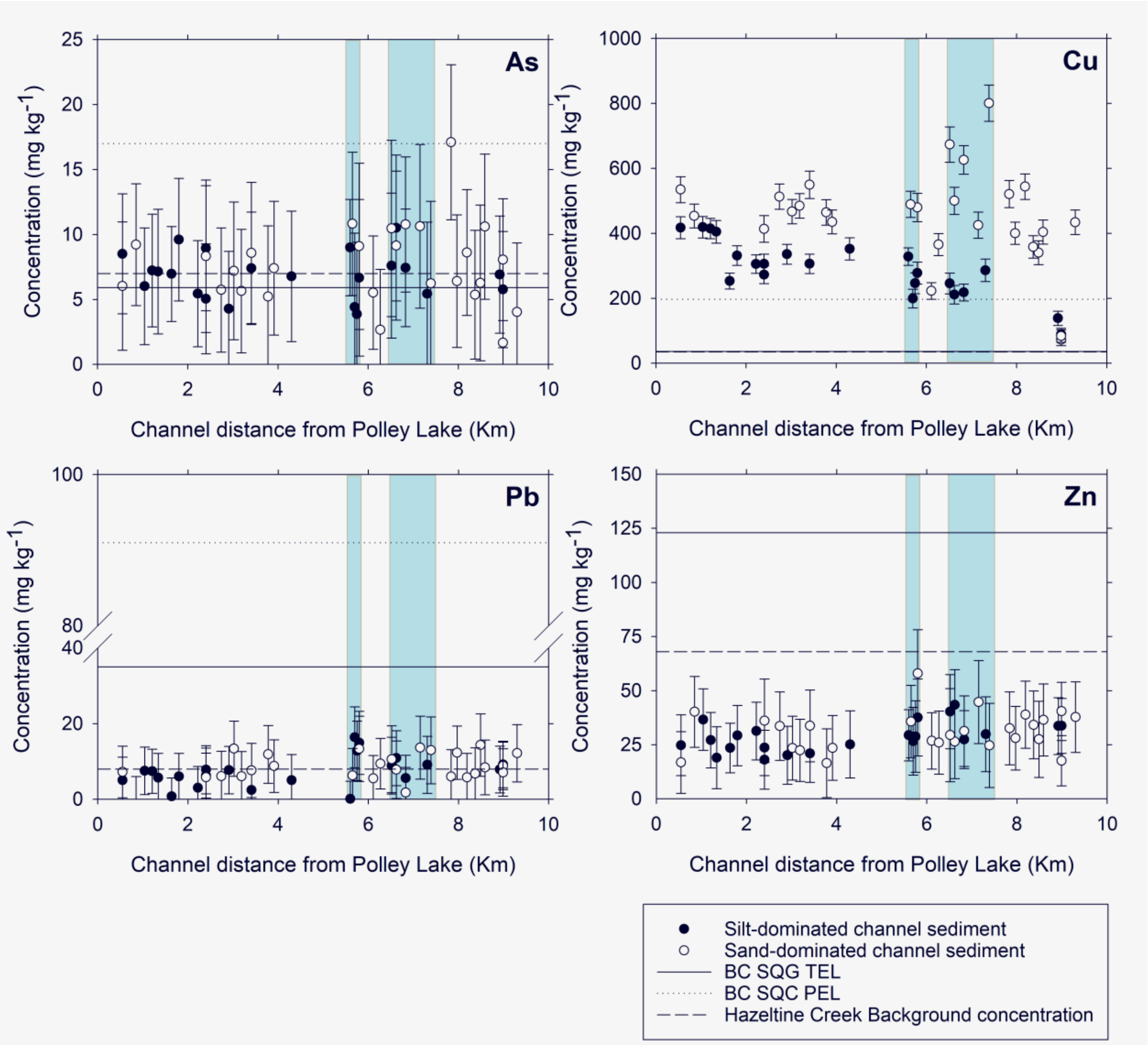

Figure 3. Metal concentrations in stream channel sediments in Hazeltine Creek. Data plotted for silt-rich and sand-rich sediments measured in the field by pXRF. Shaded areas represent the location of two bedrock gorges through which Hazeltine Creek flows. British Columbia (BC) Sediment Quality Guidelines (SQG) and background concentrations are also plotted. Note: the BC SQC PEL for Zn (315 mg kg^{-1}) is not plotted. Note: for Cu, the BC SQC TEL (36 mg kg^{-1}) and background concentration (35 mg kg^{-1}) are very similar and overlap.

Table 3. British Columbia Sediment Quality Guideline concentrations (mg kg^{-1}) for selected metals and the metalloid As (BC Ministry of Environment, 2017).

	Threshold Effect Level (TEL)	Probable Effect Level (PEL)
As	5.9	17
Cd	0.6	3.5
Cu	35.7	197
Pb	35	91
Zn	123	315

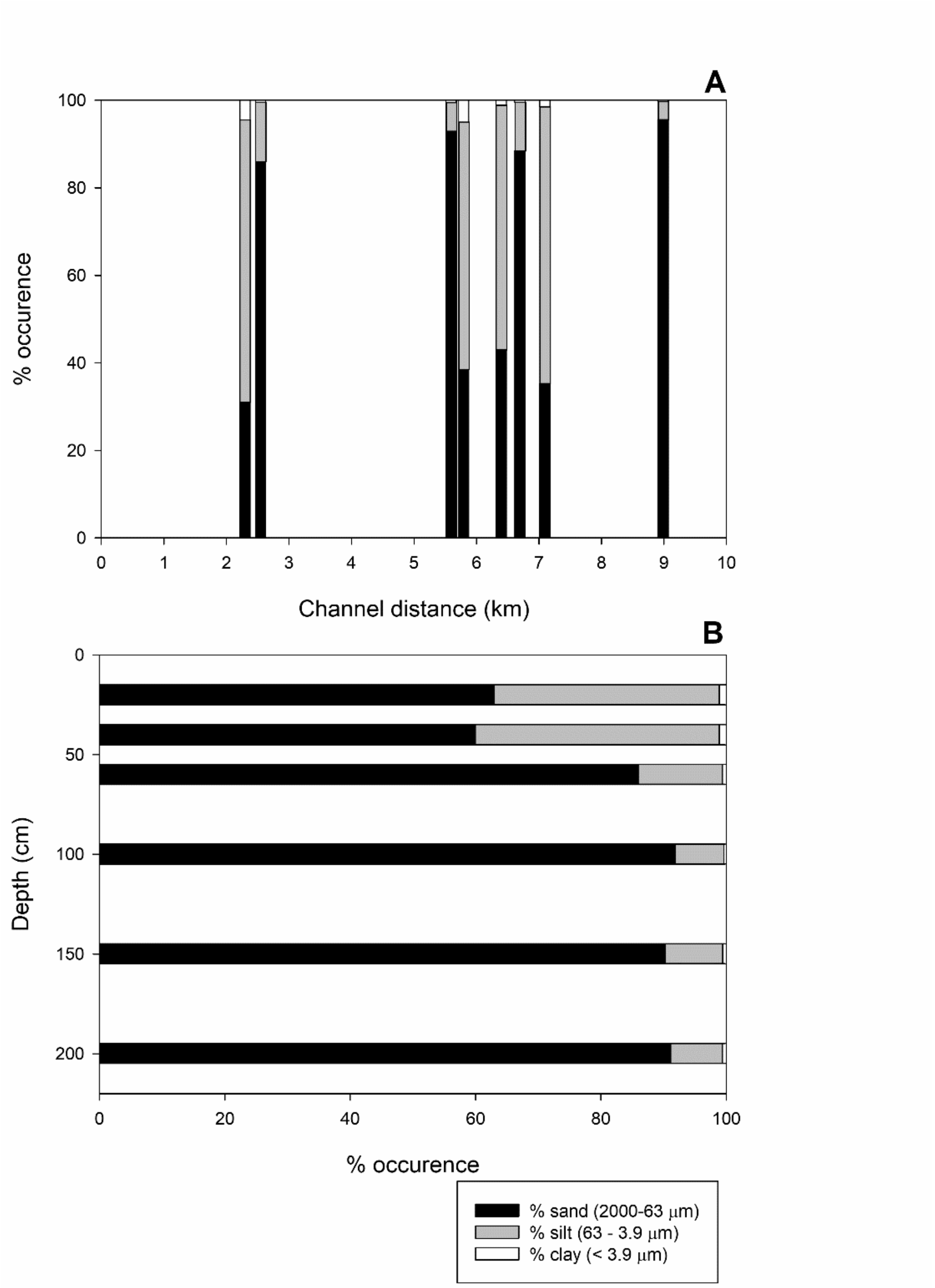

Figure 4. Percentage occurrence of sand (2 mm – 63 µm), silt (63 – 3.9 µm) and clay (< 3.9 µm) in a selection of river channel sediments (A) and floodplain sediment at site MP26 (B)

Figure 3 also indicates that for Cu, which is present in concentrations above the BC PEL, there is a general downstream trend of reducing concentrations in the silt-rich channel sediments (r^2 = 0.68). This is likely to reflect hydraulic sorting and/or dilution of Cu in these silt-rich sediments (c.f. Lewin and Macklin, 1987). The same downstream pattern does not exist for Cu concentrations in sand-rich sediments (r^2 = 0.08). It is apparent that the weaker downstream relationship for Cu in sand-rich sediment is influenced by the presence of higher Cu concentrations present in material sampled within two bedrock gorges in the lower half of Hazeltine Creek (Figure 3).

The spatial trends observed in Cu concentrations in silt-rich and sand-rich channel sediments may reflect the influence of hydraulic sorting driven by discharge and associated stream power variation during the spill event. Finer-grained, silt-rich materials will have been preferentially transported in the earlier and later stages of the spill-associated event, during the rising and falling limbs of the flood. Transport of sand-rich material would have been highest during peak flow, which would also have reworked finer-grained material released earlier in the event. The preferential transport of fine-grained, silt-rich material during the falling limb of the flood event explains the 'top-dressing' of sediments with a higher proportion of silt over coarser material on the floodplain. For example, at site MP26, floodplain sediment at 0-40 cm depth contains on average 38% silt, compared to an average of 10% silt in sample below (Figure 4b).

The steeper, confined bedrock gorges in the lower half of Hazeltine Creek will have seen the highest stream powers during the flood event, which would have resulted in the winnowing of the silt-rich sediment fraction, notably during the falling limb of the flood event. This will have left stream sediments in those reaches relatively rich in sand-rich material, shown to contain higher Cu concentrations in both the stored tailings (Figure 2) (SRK Consulting (Canada) Inc., 2015) and channel sediments (Figure 3). The sand-rich sediments sampled in the bedrock gorges contain the highest Cu concentrations in channel sediments measured along Hazeltine Creek, with Cu concentrations in sand-rich sediments being lower at sample sites between the two gorges. This results in the previously noted lack of a clear distance-concentration relationship for Cu in sand-rich sediments. The influence of stream power and similar grain-size effects in influencing distance-concentration relationships has been previously noted by Graf (1990) following the Church Rock tailings spill. These results indicate the potential complex geomorphologically-, hydraulically- and sedimentologically-influenced controls on the dispersal and storage of material released by the tailings spill and on subsequent spatial trends in channel sediment metal concentrations.

To further evaluate the impact of the spill on channel sediment geochemistry, metal concentrations determined in 2014 (immediately post-spill) and 2015 (one year after the spill) are plotted alongside concentrations measured in the 9 years prior to the dam failure from samples collected at two sites, one in the upper, and one in the lower reaches of Hazeltine Creek (Figure 5). Sample site locations are shown in Figure 1. Concentrations of metals and As within river channel sediments indicate that Cu and V showed the largest enhancement compared to pre-spill levels in Hazeltine Creek . For example, maximum post-spill Cu concentrations are 17 and 19 times more enriched in the upper and lower Hazeltine Creek, respectively. In comparison, the maximum enrichment for As (2.3 times), Pb (2 times) and Zn (1.2 times) are much lower. The concentrations measured in samples collected in 2014 and 2015 indicate that the upper reaches of Hazeltine Creek were more affected by the spill. Concentrations of Cu reduced from 952 to 512 mg kg-1 at the upper site (Figure 5), but concentrations at the lower site had reduced to 88 mg kg^{-1} in 2015 compared to 556 mg kg^{-1} in 2014 (Figure 5). The concentrations present in samples collected after the spill, and differences between upper and lower Hazeltine Creek, reflect, at least in part, the influence of grain-size effects and hydraulic sorting noted previously. However, these patterns are also to likely reflect the spatially-variable nature of post-spill remediation works. For example, in the lower part of Hazeltine Creek (upstream of the lower Hazeltine Creek sample site), it potentially reflects the influence of settling ponds, constructed. Copper concentrations in the lower Hazeltine Creek sample site (downstream of the settling pond) in 2015, were 89 % (silt-rich sediment) and 84 % (sand-rich sediment) lower than those measured at the same location in 2014, after the spill but prior to the ponds' construction (Figure 5).

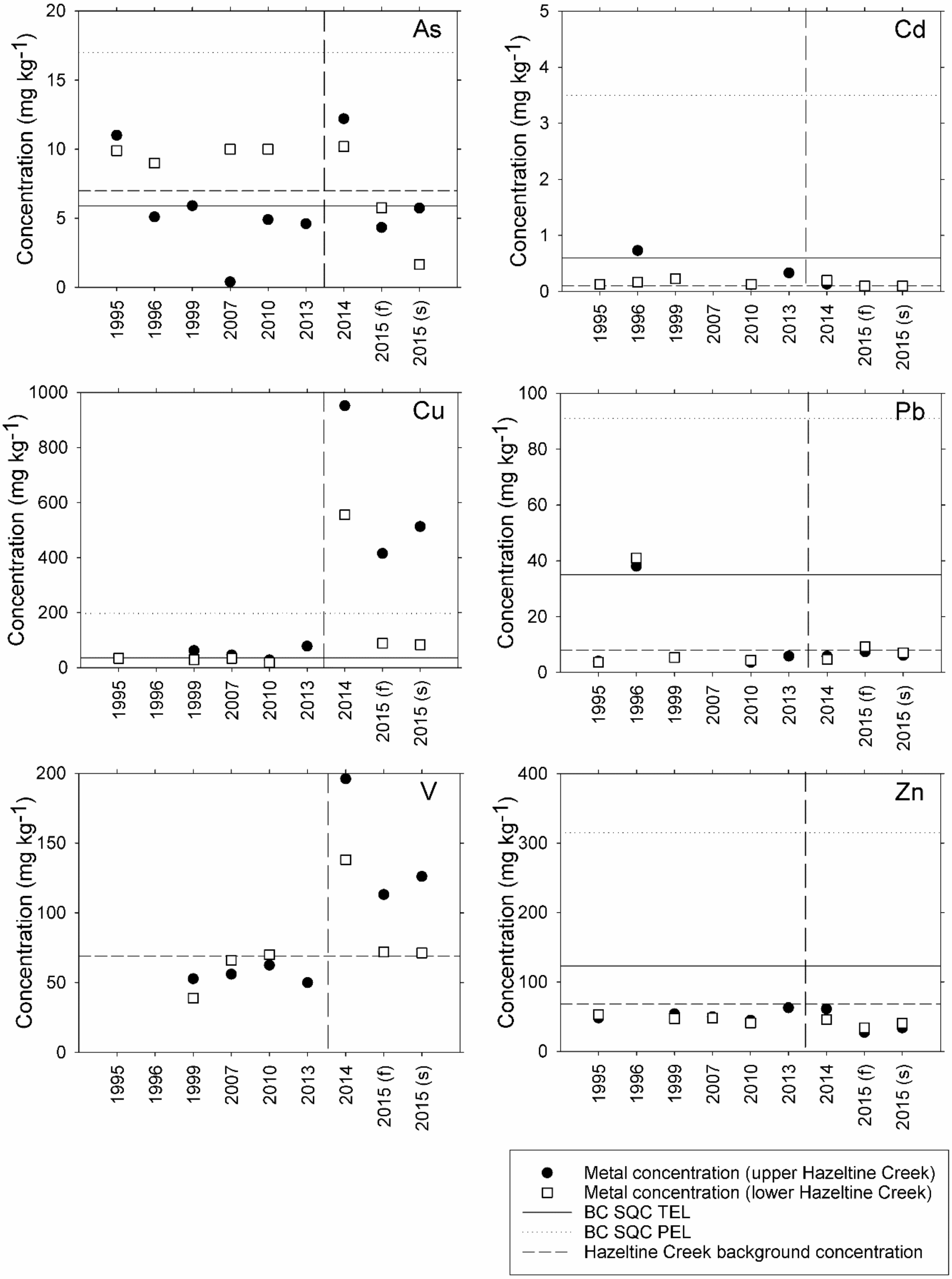

Figure 5. Metal and As concentrations pre- and post-spill in Hazeltine Creek channel sediments. The upper and lower Hazeltine Creek data sampled from locations equating to sites MP32 and MP72, respectively. Data for 1995-2013 from Minnow Environmental Inc. (2015), data for 2014 from SRK Consulting (Canada) Inc. (2015) and for 2015 from this study. Data for 2015 is provided for both fine-grained silt rich (f) and coarser-grained, sand-rich (s) sediments. Metal and As concentrations

determined following aqua regia digestion. The vertical dashed line separates pre- and post-spill samples. Note: for Cu, the BC SQC TEL (36 mg kg^{-1}) and background concentration (35 mg kg^{-1}) are very similar and overlap.

Metals in floodplain sediments

Metal concentrations in exposed floodplain profiles at sites MP14, MP25 and MP75 show pre-spill stream sediments overlain by between 0.9 – 1.6 m of Cu- and V-rich material released by the spill (Figure 6). At site MP26, the Cu- and V-rich spilled material is more than 2 m thick. The thickness of metal-rich material deposited and currently stored on the floodplain following the spill is spatially heterogeneous; a pattern that was also recorded following the 1998 Aznalcóllar tailings spill in Spain (Gallart et al., 1999). Concentrations of Cu are generally above the BC SQG PEL in the upper meter of these deposits with concentrations ranging from 560 to 1550 mg kg^{-1}. With the exception of site MP26, from c. 1 m below ground level, Cu concentrations are lower (40-581 mg kg^{-1}), below the BC SQG PEL, and reflect the background Cu concentration determined for Hazeltine Creek (Table 2). Similarly, site MP26 excepted, V concentrations in floodplain sediments deeper than 1m below ground level (75-180 mg kg^{-1}) are also similar to the upper range of background concentrations, whereas concentrations above this depth are higher (120-235 mg kg^{-1}). Arsenic concentrations (Supplementary Material 2) display a similar down-profile pattern to Cu and V, and although concentrations in the upper meter are above the BC SQG TEL, all concentrations with the exception of one, fall below the BC SQG PEL. Lead and Zn are below BC SQG TEL concentrations in all samples (Supplementary Material 2), and display no clear down-profile trends.

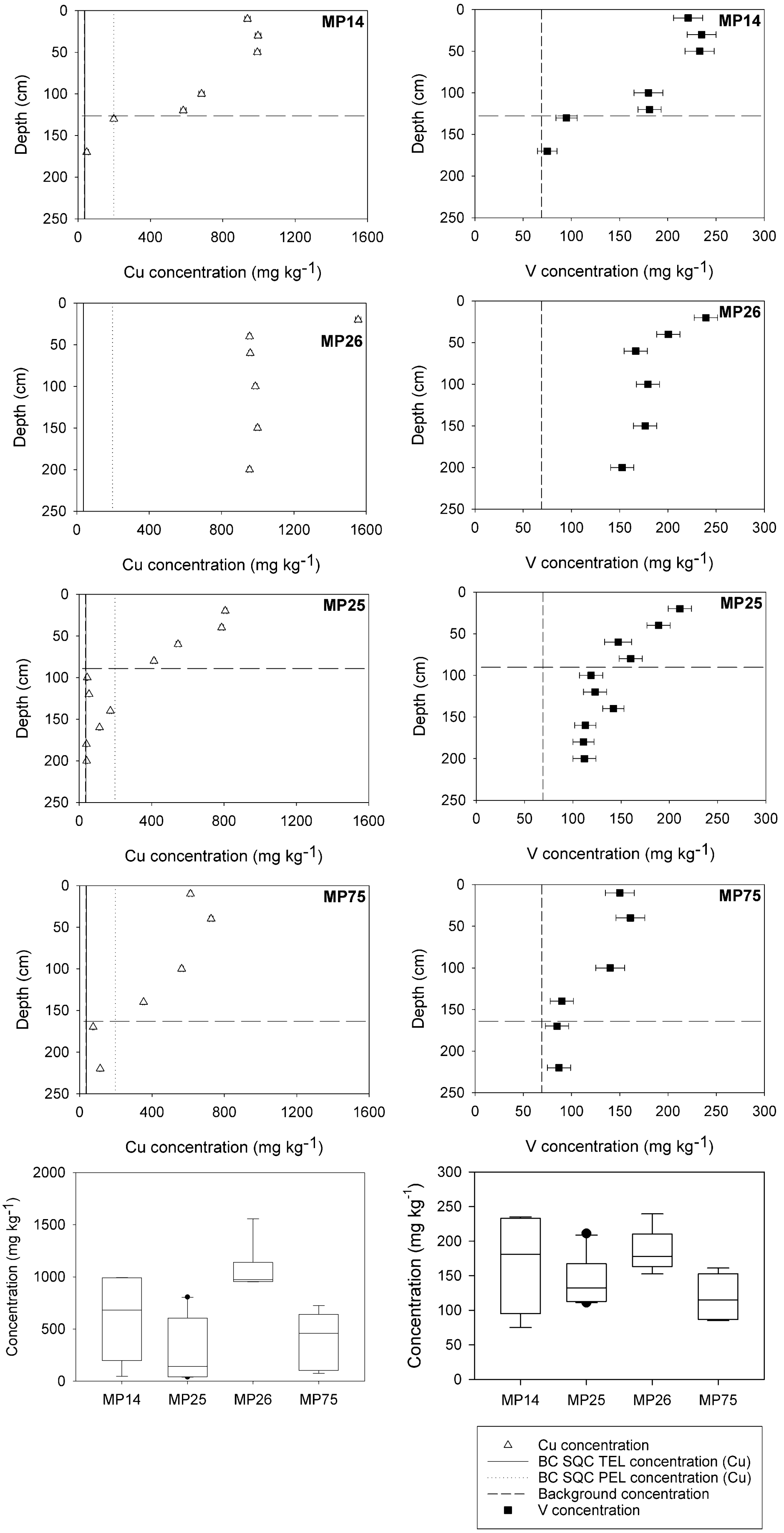
MP14
MP26
MP25
MP75
Depth (cm)
0
50
100
150
200
250
Cu concentration (mg kg-1)
0
400
800
1200
1600
V concentration (mg kg-1)
0
50
100
150
200
250
300
Concentration (mg kg-1)
2000
1500
1000
500
0
MP14
MP25
MP26
MP75
△ Cu concentration
BC SQC TEL concentration (Cu)
BC SQC PEL concentration (Cu)
Background concentration
■ V concentration

Figure 6. Copper and V concentrations from four floodplain profiles in Hazeltine Creek. Concentrations determined by ICP-MS following aqua regia digestion. British Columbia (BC) Sediment Quality Guidelines (SQG) and background concentrations are also plotted. Note: no SQC has been determined for V, and for Cu, the BC SQC TEL (36 mg kg^{-1}) and background concentration (35 mg kg^{-1}) are very similar and overlap. The horizontal line denotes the boundary between spill material and the pre-spill floodplain material (note all samples at site MP26 were within spill material). Cu and V concentrations in the four profiles are also compared using a boxplot.

Concentrations of Cu and V in the floodplain profiles at site MP75, in lower Hazeltine Creek (c. 8.5 km channel distance), are generally lower than those at site MP14, in upper Hazeltine Creek (c. 1.5 km channel distance). Median and peak Cu concentrations are 682 mg kg^{-1} and 990 mg kg^{-1}, respectively at site MP14, and 458 mg kg^{-1} and 730 mg kg^{-1} at site MP75. However, the floodplain at site MP26 (c. 6 km channel distance), approximately half-way between Polley Lake and Quesnel Lake, contains the highest concentrations of Cu and V (median and peak Cu concentrations are 970 mg kg^{-1} and 1550 mg kg^{1}, respectively). This suggests that, although there is a general down-profile reduction in Cu and V concentrations at most sites, there is not a simple down-stream trend of generally reducing metal concentrations in the floodplain deposits. Indeed, highest concentrations occur in material deposited in the middle part of the study reach. This may be related to the bedrock gorge reaches between 5.5 and 8 km channel distance (Figure 3) creating a backwater effect and enhancing floodplain sedimentation, and the deposition of metal-rich material immediately upstream.

River sediment provenance

Lead isotope signatures have been used to establish the provenance of river channel and floodplain sediments in Hazeltine Creek and to quantify the contribution from key sediment sources (Table 1) within the catchment to these sediments following the tailings spill.

Ratios for ^{207}Pb/^{206}Pb and ^{208}Pb/^{206}Pb in these potential source materials form a linear mixing linear trend with the signatures for sand-rich tailings and background sediments (indicative of geogenic Pb isotope signatures) forming the end members (Figure 7). Native channel sediments that pre-date the spill, silt and clay-rich tailings and mixed tailings plot between these end-members (Figure 7). The signatures for river channel and floodplain sediments deposited by the spill plot between the end-members at varying points along the linear trend and show that they are derived from a mixture of these source materials. Similar trends are also apparent in plots for ^{208}Pb/^{204}Pb, ^{207}Pb/^{204}Pb and ^{206}Pb/^{204}Pb (Supplementary Material 3).

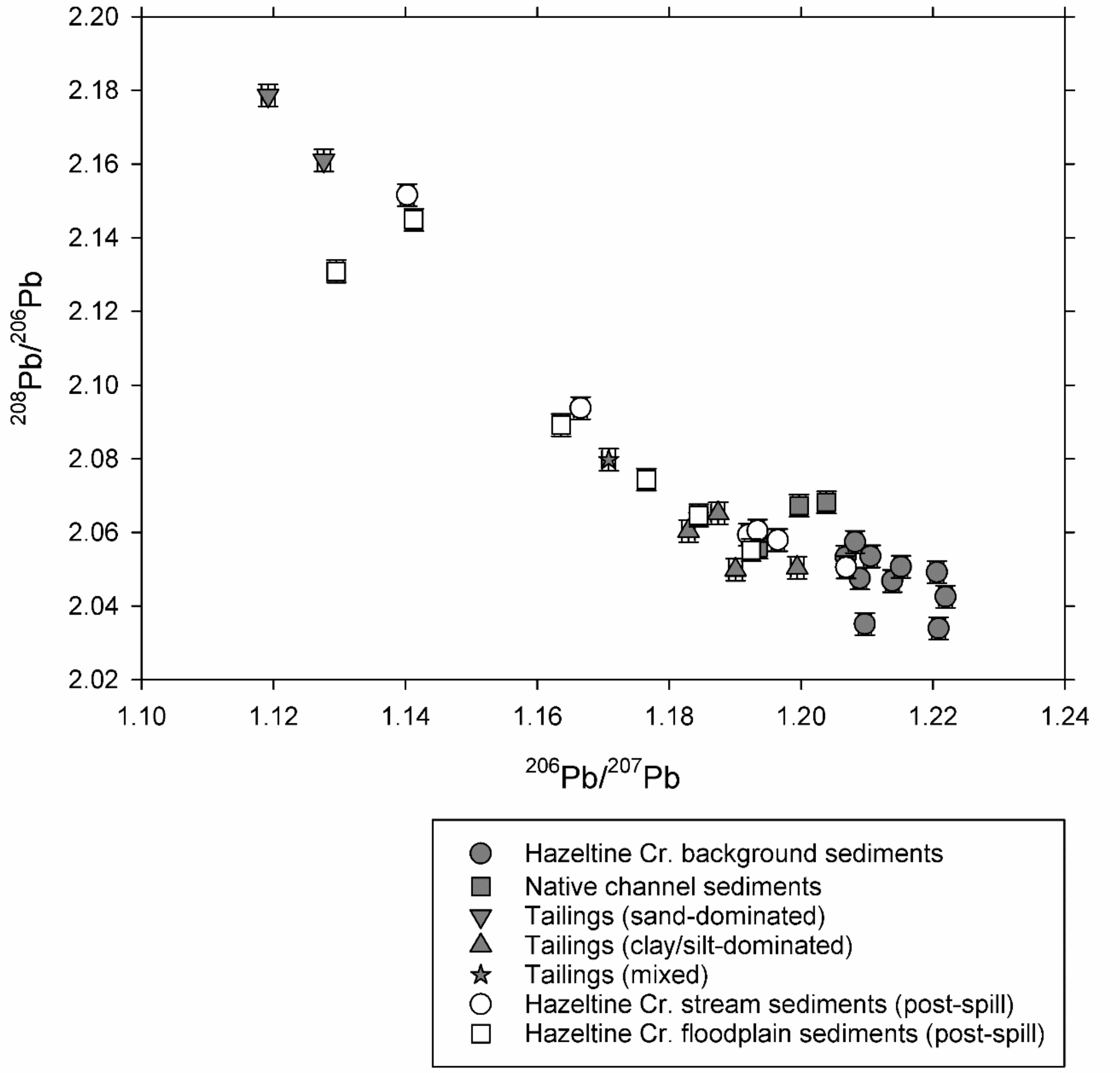

Figure 7. Ratios of ^{206}Pb/^{207}Pb and ^{208}Pb/^{206}Pb determined in river channel and floodplain sediments and tailings material from Hazeltine Creek.

The proportion of river channel sediment presently in Hazeltine Creek sourced from native channel sediments and mixed tailings is very low (both on average <2%) across all sample sites for which the mixing model was run (Figure 8a). However, the proportion of river channel sediment sourced from background sediments unaffected by mining, and sand-rich and silt/clay-rich tailings is greater, but also spatially variable. This indicates that the river channel sediments present within Hazeltine Creek following the spill reflect a mixture of background sediment (average = 27%), eroded from the valley floor during and following the accident, and tailings (on average, 28% sand-rich and 42% clay/silt-rich) released by the accident. River channel sediment samples with higher Cu concentrations (samples

MP38b, MP49, MP55b [430-670 mg kg^{-1}]) contain a greater proportion (34-80%) of sand-rich tailings released by the spill (Figure 8a). Samples collected from approximately 6.5-7 km channel distance (samples MP38a, MP55a, MP55b and MP59b) are primarily composed of clay/silt-rich tailings (52-90%) and have generally lower, but still enriched, Cu concentrations (200-270 mg kg^{-1}).

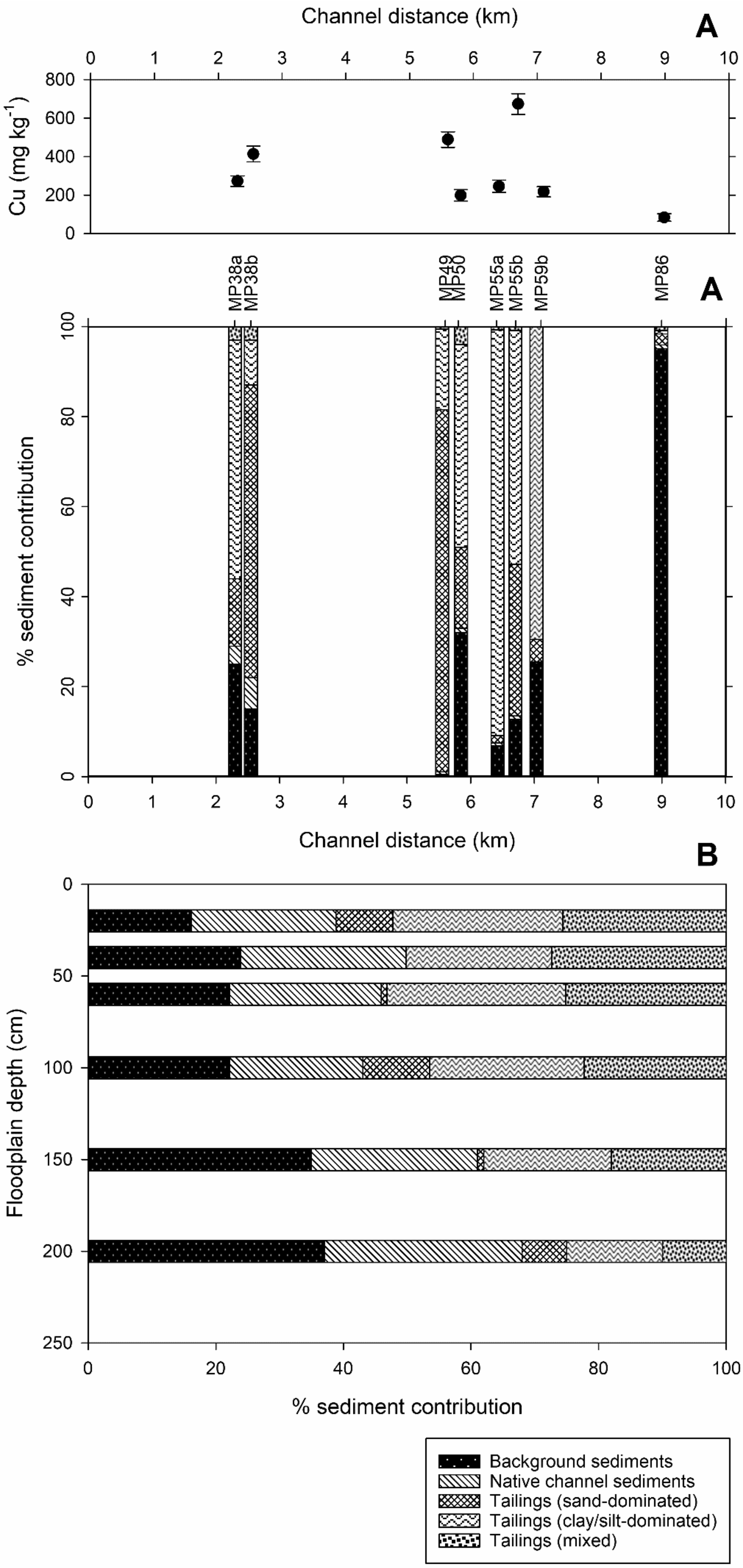
A
Channel distance (km)
0
1
2
3
4
5
6
7
8
9
10
Cu (mg kg^{-1})
800
600
400
200
0
MP38a
MP38b
MP49
MP50
MP55a
MP55b
MP59b
MP86
A
% sediment contribution
100
80
60
40
20
0
Channel distance (km)
B
Floodplain depth (cm)
0
50
100
150
200
250
% sediment contribution
0
20
40
60
80
100
Background sediments
Native channel sediments
Tailings (sand-dominated)
Tailings (clay/silt-dominated)
Tailings (mixed)

Figure 8. Cu concentrations and percentage sediment contribution in stream sediments (A) and percentage sediment contribution in floodplain sediment at site MP26 (B).

At the end of the study reach, river channel sediments at site MP86, draining out of the second of two settling ponds that were constructed post-spill to reduce sediment fluxes to Quesnel Lake, contained Cu, Pb and Zn at levels below the respective BC SQG PEL concentrations. This material comprised 95% background sediments and indicates the success of the settling ponds in trapping sediments, and particularly tailings-rich material. This is in notable contrast to sites upstream of the settling ponds which contain channel sediments estimated to be composed of 67-99% of tailings material (of any type), and therefore associated with mining-related sources.

With respect to floodplain material (Figure 8b), it is evident that samples up to 100 cm depth contain a greater proportion of tailings (50-61%) than samples at 150 cm (39%) or 200 cm (32%). Material at 150 and 200 cm depth contains a greater proportion of background and native channel sediments (up to 68 % combined) compared to the upper 100 cm (up to 49% combined). The fingerprinting indicates that this material deposited on the floodplain contains a mixture of released tailings and material eroded by the flood wave that resulted from the TSF failure (Figure 8b). The downprofile changes in the relative proportions of material derived from mining and non-mining sources indicates that the proportion of these materials varied during deposition.

Concentrations in material at site MP26 are above guideline and background values, especially in the upper profile (Figure 6). However, given that 38-68% of this material is derived from non-mining sources, this highlights that there has potentially been a degree of physical dilution of mine waste by large-scale erosion of ‘clean’ valley floor soil and sediment during the spill, and common during exceptionally large tailings dam failures (Macklin et al., 2006). This is especially true in cases similar to Mount Polley, where metal and As concentrations are low in the unmineralised parts of catchment and where there have been no historical mining or metallurgical activities resulting in watercourse contamination prior to the construction of the TSF (see Macklin et al., 2006).

Implications for management of river systems impacted by TSF failures

Previous studies have sought to compile chronologies of TSF failures, to analyse the spatial and temporal trends in occurrence (Martin and Davies, 2000; Rico et al., 2008), and the cause of failures (e.g. Lyu et al., 2019). From these studies, and from information held in databases such as WISE (2020), it is evident that the magnitude of impact, both environmentally and socio-economically are very varied. It is also apparent that there is a lack of consistent data collected on TSF failures. Each failure is unique, in terms of the volume and composition of spilled material and the physical environmental setting into which that material is released (Kossoff et al., 2014). For example, work by Rico et al. (2007) demonstrated a correlation ($r^2 = 0.56$) between the volume of spilled material and the run-out distance of that material for 28 TSF failures. However, as the authors note, the scatter within the data demonstrates the importance of the characteristics of the spill and the topography of the recipient environment. Of particular note in influencing the dispersal, storage and longer-term fate of spilled tailings will be the geomorphology of the recipient river system (Macklin et al., 2006; Kossoff et al., 2014). Figure 9 plots the relationship between volume of spilled material and run out distance for the 28 TSF failures included in Rico et al.'s (2007) study, plus an additional 16 failures for which the data are available in the WISE (2020) database, including the Mount Polley event (Supplementary Material 4). It is apparent that the analysis of the larger number of events reduces the strength of the correlation relation ($r^2 = 0.25$), further highlighting the influence of event-specific characteristics. It is also apparent that the Mount Polley event has a relatively low run-out distance in relation to the volume of tailings released (Figure 9), determined by the relatively short distance between the TSF failure and Quesnel Lake.

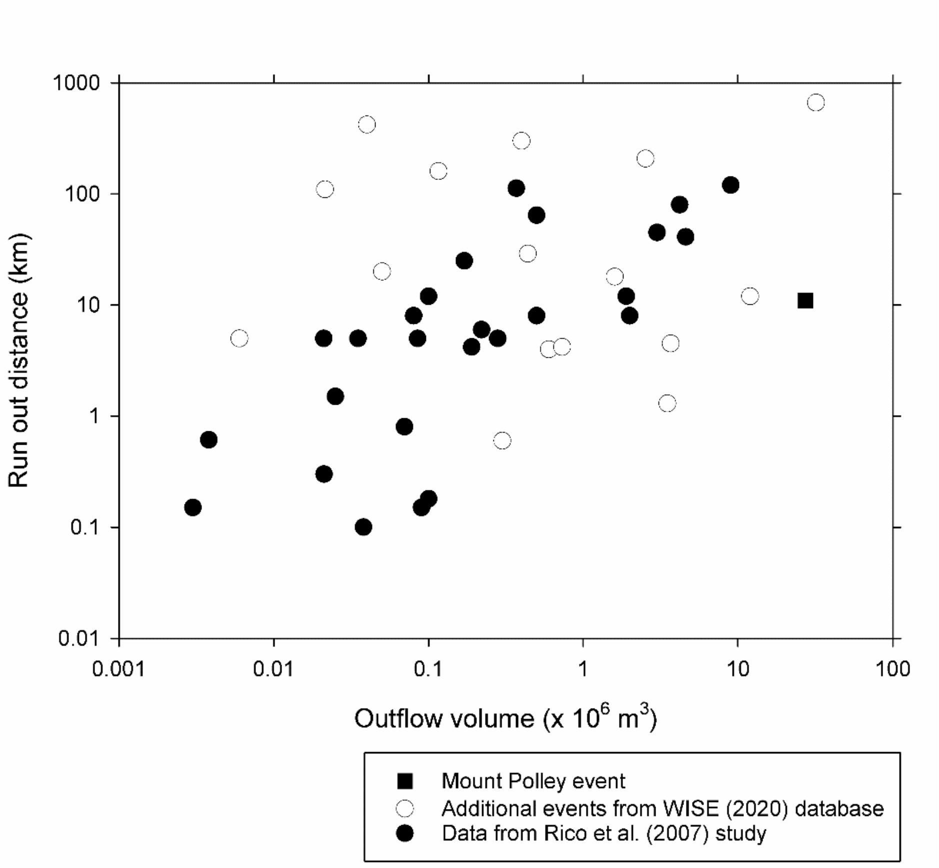

Figure 9. Relationship between the volume of spilled material and run out distance for 44 TSF failures 1965-2020.

This study has demonstrated substantial contribution of eroded catchment materials to the volume of material deposited within river systems following a tailings spill. Therefore, consideration of the potential geochemical and geomorphological disruption within recipient river systems needs to factor in the potential influence of these erosion process that may be initiated by the spill, but will themselves be influenced by the characteristics of the spill event (e.g. water volume, flow, tailings load). The initiation of erosive processes, as exemplified at Mount Polley, will also influence the volume of particulate material (tailings and eroded sediments) that are deposited within river systems and may need to be handled as part of remediation or reclamation works.

In the case of the Mount Polley TSF failure, although non-mining sediments helped to reduce overall metal and As concentrations, the physical impacts of the spill on Hazeltine Creek and Quesnel Lake were substantially amplified as a consequence of the flood wave eroding and remobilising very large volumes of pre-mining valley floor and tributary deposits. So while the concentrations of metals and As released into Hazeltine Creek were significantly smaller than in some other recent TSF failures (Bird et al., 2008; Macklin et al., 2003), the scale of habitat, river and lake environment damage was very substantial indeed. If unprecedented rates of sediment delivery to a catchment system by mining activity is considered to be an act of pollution, and the destruction of pristine river ecosystems in the 21st century viewed as being unacceptable, then the Mount Polley TSF failure represents one of North America’s most significant recent environmental disasters.

CONCLUSIONS

Analysis of river channel sediments following the Mount Polley tailing spill show concentrations of Cu in Hazeltine Creek exceed the British Columbia Sediment Quality Guideline Probable Effect Level. Concentrations were found to be highest in coarser-grained, sand-rich sediments and tailings. Spatial trends in metal concentrations in sand-rich and clay/silt-rich river channel sediments reflect the influence of hydraulic sorting, notably the differential transport and deposition of finer and coarser material on the rising and falling limbs of flood wave caused by the TSF failure. Deposition of material on the floodplain of Hazeltine Creek resulted in 1-2 m of Cu- and V-rich material burying the former floodplain surface. Lead isotope analysis and multivariate mixing modelling indicated that river channel sediments predominantly comprise a mixture of released tailings and catchment soils and sediments eroded by the flood wave. The dominance of spilled tailings was also seen in the material deposited on the floodplain surface, but up to 50% of material was derived from the erosion of catchment soils and sediments. The fingerprinting of river channel and floodplain sediments highlights the importance of erosive processes caused by TSF failures and of the contribution of eroded catchment soils and sediments to the sediment loads transported and deposited following the failure. These data indicate that the response to tailings spills by mining companies and/or governments needs to consider the volumes and composition of spilled and eroded material in the strategy.

ACKNOWLEDGEMENTS

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SUPPLEMENTARY MATERIAL 1

Table 1. Analytical quality control data for aqua regia analyses.

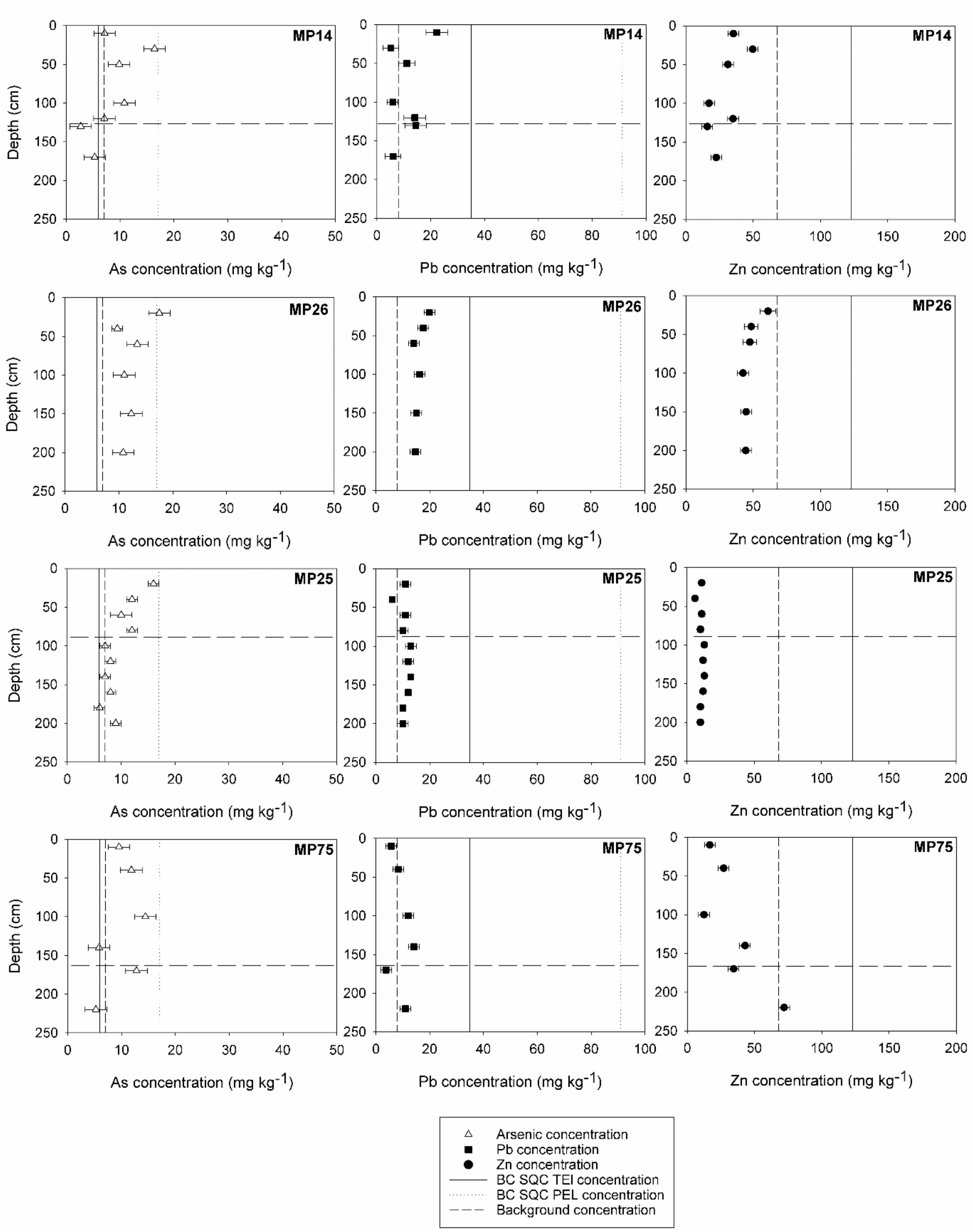
	GSD-6 Certified[a] (mg kg^{-1})	Analysed (mg kg^{-1})	% Precision[b]	Duplicate analyses[c]
As	13.6 ± 1.0	12.4	9.5	2.1-32.8 (18.3)
Cd	0.43 ± 0.03	0.36	16.2	1.1-56.7 (27.7)
Cu	383 ± 12	350	8.4	1.1-23.1 (9.3)
Pb	27 ± 4	29	5.2	3.4-9.5 (9.7)
V	142 ± 8	131	7.8	1.6-28.8 (15.2)
Zn	144 ± 7	130	6.9	2.1-32.8 (18.3)

[a] Certified values are available from: https://www.ncrm.org.cn/English/CRM/pdf/GBW07302_20160301_134249108_1713109.pdf

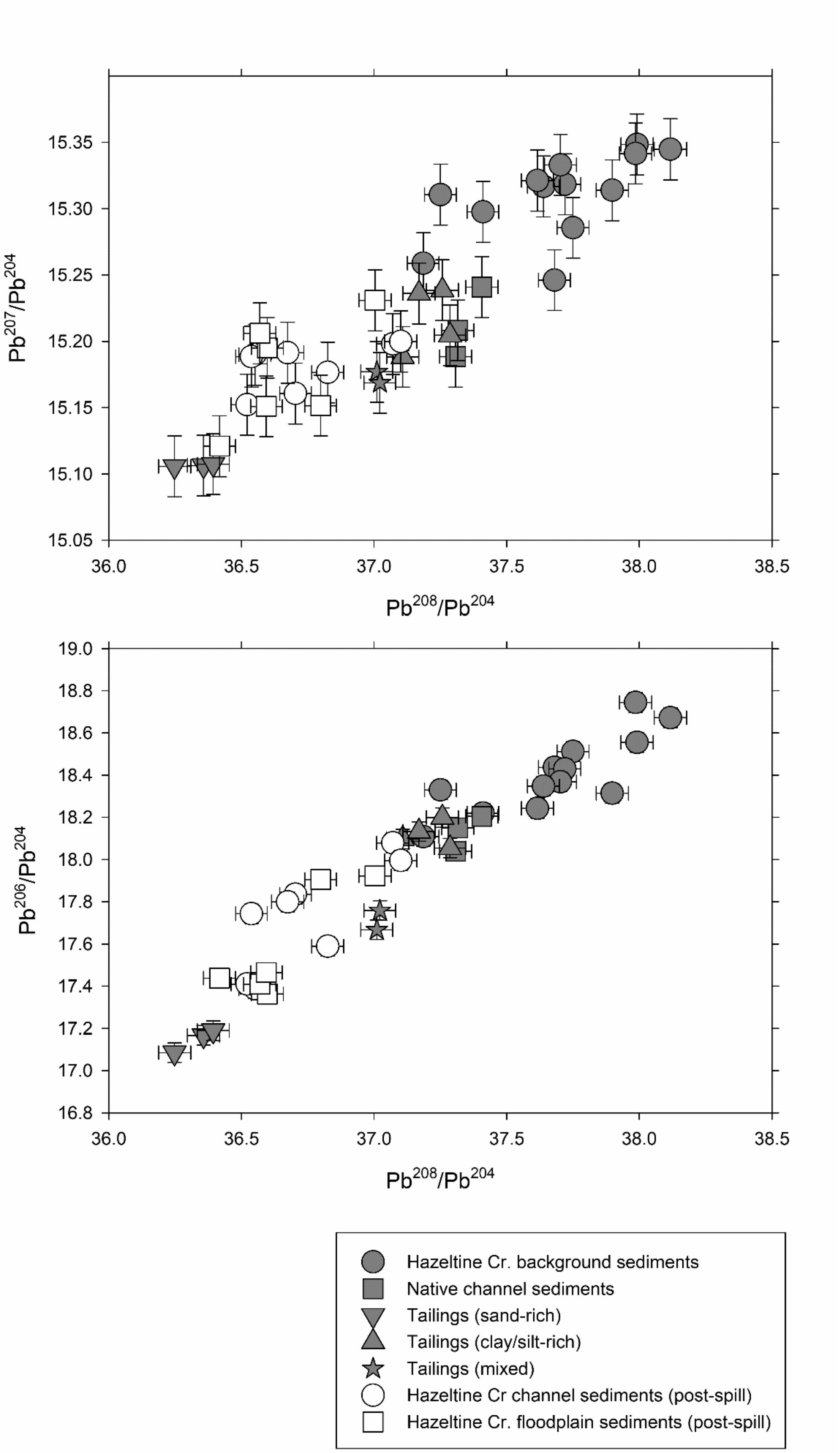
[b] Determined from replicate analysis (n = 10) of the GSD-6 CRM.

[c] Analyses to monitor the comparability of datasets provided by the Mount Polley Mining Corporation and those generated by this study. Twenty randomly selected samples previously analysed by the Mount Polley Mining Corporation, were reanalysed by this study. Data presented are the range of percentage differences between the analysis of duplicates, with mean in parentheses.

SUPPLEMENTARY MATERIAL 2



SUPPLEMENTARY MATERIAL 3



SUPPLEMENTARY MATERIAL 4

Table 1. Data for volume of material released (tailings and water) and run out distance for TSF failures 1965-2020. Data from Rico et al. (2007) are italicized and data for other events (including Mount Polley [bold] are taken from WISE (2020).

Event, Location	Date	Volume of material released (x10^6 m^3)	Run out distance (km)
San José de Los Manzanos, Mexico	2020	0.006	5
Tieli, China	2020	2.53	208
Córrego de Feijão, Brazil	2019	12	12
Cieneguita , Mexico	2018	0.439	29
Germano, Brazil	2015	32	663
Mount Polley, Canada	**2014**	**27.4**	**11**
Buenavista del Cobre, Mexico	2014	0.04	420
Huancavelica, Peru	2010	0.02142	110
Cerro Negro, Chile	2003	0.05	20
Sasa Mine, Macedonia	2003	0.1	12
Amatista, Nazca, Peru	1996	0.3	0.6
El Porco, Bolivia	1996	0.4	300
Marcopper, Philippines	1996	1.6	18
Harmony, Merriespruit, South Africa	1994	0.6	4
Niujiaolong, China	1985	0.73	4.2
Balka Chuficheva, Russia	1981	3.5	1.3
Silverton, USA	1975	0.116	160
Huogudu, China	1962	3.68	4.5
Baia Mare, Romania	*2000*	*0.1*	*0.18*
Los Frailes, Spain	*1998*	*4.6*	*41*
Omai, Guyana	*1995*	*4.2*	*80*
Stancil, USA	*1989*	*0.038*	*0.1*
Itabirito, Brazil	*1986*	*0.1*	*12*
Stava, Italy	*1985*	*0.19*	*4.2*
Cerro Negro No.4, Chile	*1985*	*0.5*	*8*
Veta del Agua N°1, Chile	*1985*	*0.28*	*5*
Ollinghouse, USA	*1985*	*0.025*	*1.5*
Phelps-Dodge, USA	*1980*	*2*	*8*
Churchrock, USA	*1979*	*0.37*	*112.6*
Mochikoshi No.1, Japan	*1978*	*0.08*	*8*
Mochikoshi No.2, Japan	*1978*	*0.003*	*0.15*
Arcturus, Zimbawe	*1978*	*0.0211*	*0.3*
Bafokeng, South Africa	*1974*	*3*	*45*
Galena Mine, USA	*1974*	*0.0038*	*0.61*
Unidentified, USA	*1973*	*0.17*	*25*
Buffalo Creek, USA	*1972*	*0.5*	*64.4*
Cities Service, USA	*1971*	*9*	*120*
Hokkaido, Japan	*1968*	*0.09*	*0.15*

Sgurigrad, Bulgaria	*1966*	*0.22*	*6*
Bellavista, Chile	*1965*	*0.07*	*0.8*
Cerro Negro No.3, Chile	*1965*	*0.085*	*5*
El Cobre Old Dam, Chile	*1965*	*1.9*	*12*
La Patagua New Dam, Chile	*1965*	*0.035*	*5*
Los Maquis, Chile	*1965*	*0.021*	*5*