Interpretation of Selenium Release from Coal Mine Waste Rock Dumps, Southeastern British Columbia, Canada

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Abstract
Teck Coal Ltd. operates five large open pit coal mines in the catchment basin of the Elk River in southeastern British Columbia. Since the late 1980's, monitoring of the river has detected increasing selenium concentrations which have been shown to originate primarily from mine waste rock. Monitoring of flow and chemistry in eleven streams receiving drainage from waste rock dumps since the late 1990's has provided an opportunity to understand selenium release from waste rock and relationships to the quantity of waste rock and seasonal factors. Drainages are uniformly basic and dominated by sulphate, calcium, magnesium and alkalinity.

Analysis of the monitoring data has shown that despite wide variations in contributing waste volume (7 to 800 million m^3), distance from the source (0 to several kilometres), and type of monitoring location (toe drain or stream), average annualized selenium release normalized to waste rock quantity in the local catchments vary over a narrow range (0.2 to 3 mg/m^3/year) and are correlated to sulphate release. The data support oxidation of pyrite as the primary origin of selenium and conservation of selenium mass in the form of selenate during leaching.

Key Words: Elk Valley, open pit, non-acidic drainage, selenium leaching

Introduction
Mining of bituminous-grade coals has occurred in the Elk Valley of south eastern British Columbia, Canada since the late nineteenth century when underground mining near the town of Fernie began (Figure 1). However, since the 1960's, coal mining has been by open pit methods from five mines in the upper reaches of the valley (Table 1). Several companies operated the mines until the operations were consolidated under Teck Coal Ltd. Coal from the mines is processed in the Elk Valley and exported by rail to a port near Vancouver, B.C. for export to off-shore markets.

Historically, water quality management for the sites has been focussed on addressing export of sediments and explosives residues from the mining areas. For example, mine permits typically contain limits for total suspended solids (TSS) and nitrogen forms (nitrate, nitrite and ammonia). Other chemical parameters have been evaluated through monitoring in some cases near the sites by the mining companies but also at a regional sampling station by the provincial government. In the mid-1980's, selenium concentrations were below the current provincial water guideline of 2 µg/L (MacDonald and Strosher 1998). An upward trend in concentrations has been apparent and in the late 1990's, average annual concentrations exceeded the water quality guideline at the Highway 93 location (Figure 1). It is apparent the upward trend in selenium concentrations is correlated with cumulative waste rock produced by mining (Figure 2) (Swanson 2010).

To investigate potential environmental effects in the watershed, the joint industry and government Elk Valley Selenium Task Force (EVSTF) was established in 1998 (Pumphrey and Gilron 2009). Initial efforts through the early 2000's and ongoing were focussed mainly on biological effects. In the mid-2000's, investigation of rock weathering mechanisms accelerated to support prediction of selenium release and development of management measures. Various studies were initiated to understand release of selenium at different scales (SRK Consulting 2008).

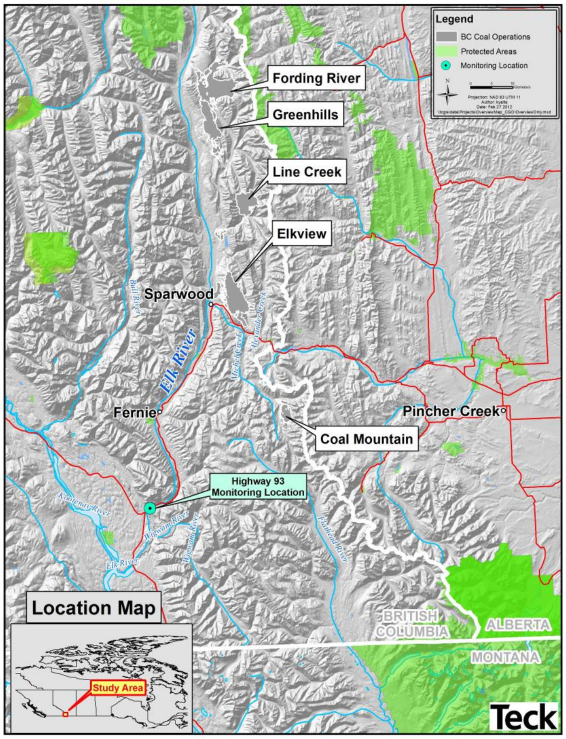


Figure 1. Location of Active Coal Mines in the Elk Valley.

Table 1. Nomenclature of Coal Mines in the Elk Valley

Mine	Abbreviation	Start of Operations
Coal Mountain Operations	CMO	1985
Elkview Operations	EVO	1969
Line Creek Operations	LCO	1982
Greenhills Operations	GHO	1982
Fording River Operations	FRO	1972

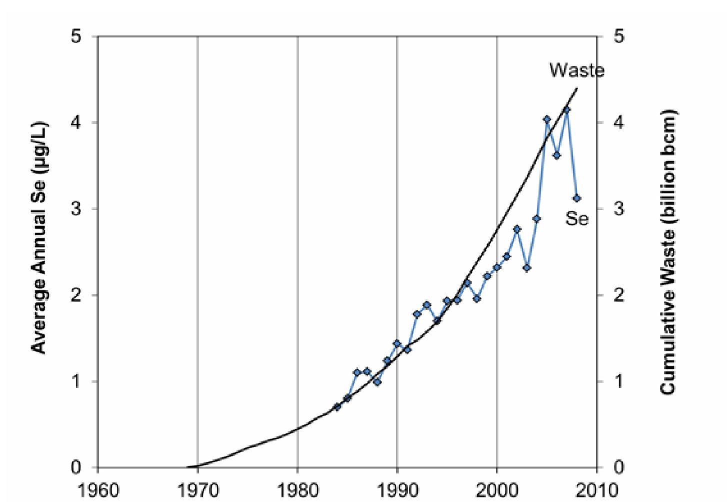


Figure 2. Trends in Selenium Concentrations and Cumulative Waste Rock Volume at all Teck Coal Operations in the Elk Valley (Swanson 2010).

This paper reports on interpretation of flow and monitoring data from full-scale facilities over several years. A companion paper (Kennedy et al. 2012, these proceedings) presents results of specific studies at Line Creek Operation (LCO) to understand selenium occurrence and release from laboratory and small-scale field experiments.

Conceptualization of Selenium Release and Leaching

The co-occurrence of selenium with sulphide minerals (SRK Consulting 2004) implies that selenium release is likely to be linked principally to weathering and leaching of sulphide minerals through the conventional oxidation of pyrite:

$$FeS_2 + 15/4O_2 + 7/2H_2O \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \qquad (1)$$

In this reaction, selenide (Se^{2-}) is expected to behave the same as sulphide (S^{2-}) and be oxidized to selenate (SeO_4^{2-}). In the presence of abundant carbonate minerals, consumption of acidity results in mildly basic conditions in weathering environments. For redox-sensitive selenium, this indicates that selenium will likely persist as selenate in solution and therefore remain highly mobile due to its low tendency to adsorb to reactive surfaces. An exception to this general conclusion may exist where locally reducing conditions develop in biofilm micro-environments (de Beer et al. 2004) allowing selenate to be converted to selenite (SeO_3^{2-}) and possibly also elemental selenium (Se^0) and selenide. The resulting solid selenium forms all reduce selenium mobility.

Selenate is highly soluble and does not readily form discrete secondary minerals analogous to the sulphate minerals (e.g. calcium selenate); however, co-precipitation of selenium with gypsum and carbonate precipitates has been reported in the literature (Fernández-González et al. 2006; Reeder et al. 1994; Staudt et al. 1994; Andara et al. 2005) and implied by sampling in the Elk Valley (SRK 2004, 2011; Kennedy et al. 2012).

The linkages between geochemical transformations that convert sulphur and selenium from low to high solubility forms, and the hydrological processes that leach the soluble minerals are not well understood. Considerations include the time required to wet-up wastes to the point at which they release water, and the effect of the seasonal flushing caused by the snowmelt event. For the purpose of this study, it is assumed that sulphate and selenate will show a similar response to hydrological processes.

The conceptual model suggests weathering and leaching of selenium should be linked to the presence of sulphide minerals, and would be correlated with sulphate leaching. Broadly, the similarity of mining methods, the extraction of coal seams from one geological formation, and relatively consistent geochemical characteristics of rocks in the Elk Valley implies that release of sulphate and selenium from waste rock should occur at similar rates from waste rock dumps at the different operations. Water quality and flow monitoring data and mining history for the operations provided an opportunity to evaluate this hypothesis and possible explanations for differences in release rates that have implications for management of selenium.

Background

Geological setting

Coal extraction occurs from the Mist Mountain Formation in the Jurassic-Cretaceous Kootenay Group. This formation consists of an inter-bedded succession of predominantly non-marine sandstone, siltstone, mudstone, rare conglomerate and thin to thick coal seams (Gibson 1985). The formation varies in thickness from 25 to 665 m and contains numerous coal seams up to 18 m thick. The coals vary in rank from low volatile bituminous to semi-anthracite in the north part of the basin. The Mist Mountain Formation is underlain by the Morrisey Formation, which is comprised of massive sandstone believed to be deposited in a coastal environment and overlain by the Elk Formation which is interpreted to have formed in a coastal fluvial-alluvial environment. The distinctive feature of the Mist Mountain Formation is that it is believed to have formed in a paleo-environment with limited marine influence.

Coal seams are exclusively extracted from the Mist Mountain Formation at most of the mines with limited mining of the Morrisey and Elk Formations. The Coal Mountain Operation (CMO) is the exception because about 30% of its waste rock comes from sandstones of the Morrisey Formation and claystones and shales of the Jurassic Fernie Group underlying the Kootenay Group.

Selenium occurrence

Various studies by provincial geologists (Ryan and Dittrick 2001; Ryan et al. 2002), the University of British Columbia (Lussier 2001) and consultants (SRK 2004; SRK 2011; Kennedy et al. 2012) have evaluated the geochemistry and mineralogy of rocks associated with coal in the Elk Valley with particular emphasis on selenium. These studies have shown that sulphur content of the rocks varies by rock type but is relatively low (about 0.2% on average). The mines in the region have no reported history of acid rock drainage (ARD) from waste rock as a result of the low sulphide content coupled with excess carbonate minerals dominated by dolomite and ankerite. Sulphur occurs as the iron sulphides pyrite and marcasite, and bound with carbonaceous materials.

Selenium concentrations show well-established correlations with rock type. Lowest concentrations of selenium and sulphur occur in sandstones (typically near 1 mg/kg) and higher concentrations in mudstones (2 to 4 mg/kg) (Ryan and Dittrick 2001; Ryan et al. 2002). Concentrations in siltstones range between these values. Maximum selenium concentrations typically do not exceed 10 mg/kg.

The mineralogical occurrence of selenium has been investigated using laser ablation by SRK (2004) and chemical techniques (SRK 2011; Kennedy et al. 2012). SRK (2004) showed that the main reactive host for selenium is iron sulphide but it was also found to be associated with other sulphur-bearing minerals (barite and gypsum). SRK (2011) showed a likely association with carbonaceous matter. Sulphur and selenium concentrations are commonly correlated confirming the overall link to the occurrence of sulphur.

Selenide minerals have been rarely identified in tailings but they do not represent a significant form of selenium (unpublished Teck Coal Ltd. research).

Climate and hydrology

The climate at Sparwood (elevation 1138 m above sea level, Figure 1) is characterized by average annual temperatures of 4.3°C with a monthly average daily range of -7.7°C (December) to 15.4°C (July) (Environment Canada 2011). Total precipitation averages 603 mm and about two-thirds of precipitation falls as rain. Stream flows in the area are dominated by peak flows resulting from snowmelt in May.

Mining method

Mining occurs at all five mines by conventional drill-and-blast and truck-and-shovel methods. Raw coals are hauled and conveyed to processing plants located at lower elevations near each mining operation. Oversize waste (referred to as coarse coal rejects or CCR) from processing is typically disposed in dedicated dumps located adjacent to the plants, and fine process wastes (tailings) are deposited in impoundments. Waste rock disposal occurs dominantly in external dumps with tipping faces varying from tens to hundreds of metres in height. Backfilling into exhausted sections of pits occurs at some locations. Historically, co-disposal of waste rock and CCR has taken place at some mines.

Methods

Site selection

To provide a consistent basis for comparison of sites within the Elk Valley, monitoring locations were identified that provided a reliable flow monitoring database, coincident water chemistry data and mining records that allowed release of selenium and sulphate to be linked to waste rock quantities. Twenty-six monitoring locations at all the operations formed the initial list of candidate sites which were eventually reduced to ten using the above criteria. Sites were eliminated for a variety of reasons including most commonly the presence of both CCR and waste rock contributing to the drainage and lack of certainty about the quantity of waste rock contributing to the monitoring location. A few locations lacked flow and/or relevant chemistry data.

Tables 2 and 3 provide some characteristics of the ten locations selected for inclusion in the study. The majority of locations are settling pond (SP) outfalls located several hundred metres to kilometres from the nearest spoil. In most cases, waste rock dumps represent a relatively small proportion of the catchment area, and the waste rock dump is not necessarily located in the headwaters of the catchment. For Henretta and Kilmarnock Creeks, monitoring occurs on a large creek that flows through waste rock. Porter, Harmer, Erickson, Six Mile and Porter Creeks are monitored distant from the nearest spoils in their headwaters. Cataract, Swift, West Line, Bodie and Corbin Creeks have relatively high proportions of waste rock in their catchment areas. None of the monitored flows are seeps.

The volume of spoil in the catchment areas varies over nearly three orders-of-magnitude from the short duration (1 year) small (7 million bank cubic metres, BCM) Six Mile waste rock dump at EVO to the active nearly billion BCM Kilmarnock Spoils at FRO (Table 3).

Data quality

The flow and waste rock quantity data used in the study were collected by several different mine operators through the late 1990's to 2009. Chemical analyses were also performed by different laboratories. For the purpose of this study, data quality was not investigated but assumed to have been evaluated by the mines' environmental departments as part of annual data collection efforts.

Data reduction

Several data reduction steps were followed with the intent of calculating annual flow and chemical loading for each monitoring location. Flow monitoring data were typically available on a daily basis whereas water chemistry data were available monthly or less frequently To create a continuous daily flow and concentration dataset, missing flow and concentration data for individual days were created by using the last date for which data were available.

The combined flow (Q) and concentration data (C_i) for parameter i were used to calculate loadings (L_i,day) for parameter i using the conventional formula relating concentration and flows:

$$L_{i,day}\ (\text{mg/day}) = Q\ (\text{m}^3/\text{day})\cdot 1000\ (\text{L/m}^3)\cdot C_i\ (\text{mg/L}) \qquad (2)$$

Monthly and annual loads (L_i,year, mg/year) were then calculated by summing the daily loads for each parameter.

Only years for which there were at least six monitoring rounds for flow and chemistry were included. This target was based mainly on having data representing the six months of open water (May to October) when loadings tends to be greatest due to snow melt and flushing.

Table 2. Drainage Monitoring Locations

Mine Operation	Creek	Monitoring Location	Location Type	Ratio of Undisturbed to Disturbed Area in Catchment[1]	Elevation m	Distance to Spoil m	Years of Record as of 2008	
							Se and SO_4	Flow, Se and SO_4
FRO	Henretta	HC1	Creek	Much greater than 1	1717	0	5	2
FRO	Kilmarnock	KC1	Creek	Much greater than 1	1617	600	5	2
GHO	Cataract	CC1	SP Outfall	Less than 1	1611	400	5	5
GHO	Porter	PC1	SP Outfall	Greater than 1	1576	1300	5	5
LCO	West Line	WLC	Rock drain	Greater than 1	1461	300	6	1
EVO	Bodie	BC1	SP Outfall	Less than 1	1160	600	8	5
EVO	Harmer	HC1	SP Outfall	Much greater than 1	1324	6800	6	6
EVO	Six Mile	SM1	SP Outfall	Much greater than 1	1160	2400	5	4
EVO	Erickson	EC1	Creek	Much greater than 1	1217	4000	9	5
CMO	Corbin	CPD	SP Outfall	Less than 1	1596	400	1	1

Table 3. Spoiling History at Drainage Monitoring Locations

Mine Operation	Creek	Monitoring Location	Spoiling History		2008 Spoil Volume $x10^6$ BCM	Years of Record as of 2008	
			Start	Finish		Se and SO_4	Flow, Se and SO_4
FRO	Henretta	HC1	1993	Active	147	5	2
FRO	Kilmarnock	KC1	1983	Active	970	5	2
GHO	Cataract	CC1	1984	Active	364	5	5
GHO	Porter	PC1	1986	Active	79	5	5
LCO	West Line	WLC	1981	1999	187	6	1
EVO	Bodie	BC1	1993	Active	263	8	5
EVO	Harmer	HC1	1969	Active	532	6	6
EVO	Six Mile	SM1	1980	1980	7	5	4
EVO	Erickson	EC1	1969	Active	365	9	5
CMO	Corbin	CPD	1995	Active	148	1	1

The load leached per volume of rock ($L'_{i,year}$, mg/year/m^3) was calculated based on the cumulative volume of rock in a year indicated by spoil placement history. The latter was based on volumes (in back cubic metres, or BCM's) removed from active pits rather than as-built surveys of waste rock dumps.

For most sites, several values of $L'_{i,year}$ were obtained. The average for each site was obtained and the range represented by the 95% confidence limit on the mean. The full range of $L'_{i,year}$ for each site is not considered to represent the underlying source release rate due to the effect of year-on-year leaching effects caused by below above average hydrological years. For example, lower loads might be expected during a relatively dry year due to lower rock contact, whereas higher loads might during a wet year due to leaching of soluble minerals accumulated during previous dry year(s).

Results

A wide range in concentrations of selenium and sulphate in the drainage waters were apparent in the monitoring data reflecting seasonal conditions and the proportion of mining disturbed areas in the their catchments (Table 2). For example, Kilmarnock Creek, which has a relatively high proportion of undisturbed areas in its catchment shows sulphate and selenium concentrations below 500 and 0.12 mg/L, respectively and a strong trend of lowest concentrations during high flow volumes as snow melts in the spring (Figure 3).

In contrast, Cataract Creek's catchment is mainly occupied by waste rock and shows sulphate and selenium concentrations up to 1650 mg/L and 0.64 mg/L, respectively (Figure 3). West Line Creek shows a similar seasonal trend to Kilmarnock Creek though with sulphate and selenium concentrations comparable to Cataract Creek. Flows and concentrations at some monitoring locations are also affected by water management activities at the mines including pit de-watering.

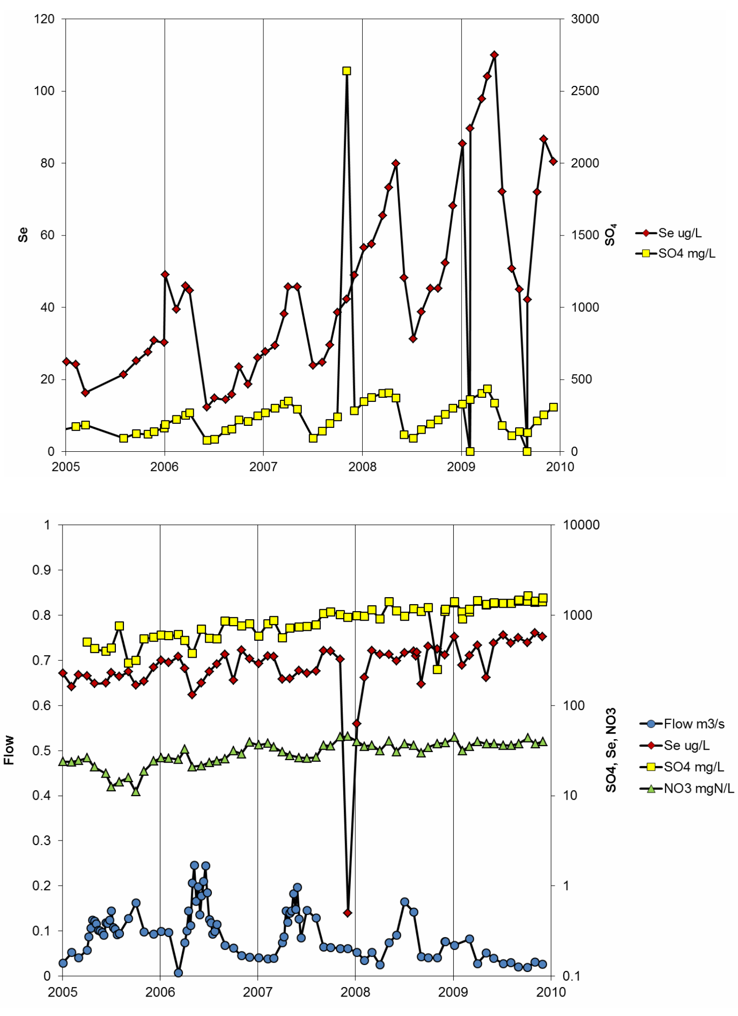
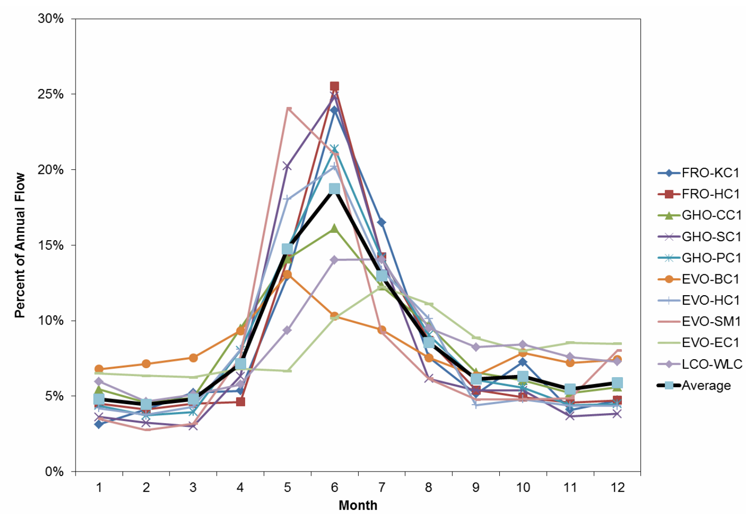


Figure 3. Monitoring Results for Kilmarnock Creek (top) and Cataract Creek (bottom).

Removal of site-specific flow effects by calculation of loadings revealed consistent seasonal trends between sites. On average, peak flows averaging nearly 20% of total annual flow occur in June and then decline through the summer (Figure 4). Some variation in when peak flows occur can be expected due to variations in elevation through the Elk Valley (Table 2) and water management measures. Sulphate (Figure 4) and selenium loads also peak coincidentally with flows though peaks tend to be more subdued and account for a lower proportion of the annual load (about 15%). As a result, concentrations tend to be lower during the freshet high flow event then increase through the summer though the response at individual sites varies, as shown in Figure 3.

Average annual selenium loads are shown in Figure 5. Error bars in the figure are 95% confidence limits calculated for the mean value. The number of years used to calculate the statistics are shown. Where only one annual value could be calculated, no error bar is shown. Average loads vary over a relatively narrow interval from a low of 0.23 mg/BCM/year for Corbin Creek at CMO to 3.3 mg/BCM/year for West Line Creek at LCO though only one annual rate was calculated. As shown, error bars overlap for most sites and there is little evidence that actual differences in loadings can be resolved.



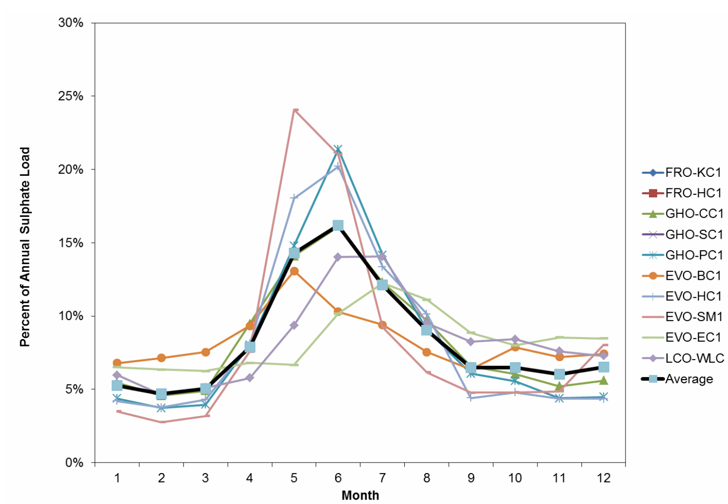


Figure 4. Average Seasonal Flow (top) and Sulphate Load (bottom) Distribution

Average annual sulphate load also varied over a narrow range (5 to 15 g/BCM/year) and correlation with selenium load was not readily apparent (Figure 6) though two of the sites at the extremes (LCO-WLC and CMO-CPD) only had one measurement resulting in uncertainty in the load. The other sites show a weak correlation of sulphate and selenium loading. Direct comparison of concentrations rather than loading allows the comparison to be expanded by removing the need for flow data. This confirms some of the differences in loadings. LCO-WLC shows a generally higher Se/SO_4 ratio than the other datasets, and particularly with references to GHO-CC1 which show similar Se and SO_4 concentrations to LCO-WLC. CMO-CPD is notable in showing much lower Se concentrations when compared to other sites with similar sulphate concentrations. Overall, an apparent downward decrease in Se/SO_4 is apparent as overall concentration decrease. With the exception of CMO-CPD, EVO-SM1 shows the lowest Se/SO_4 in the dataset and explains some of the scatter in the relationship between annual loading.

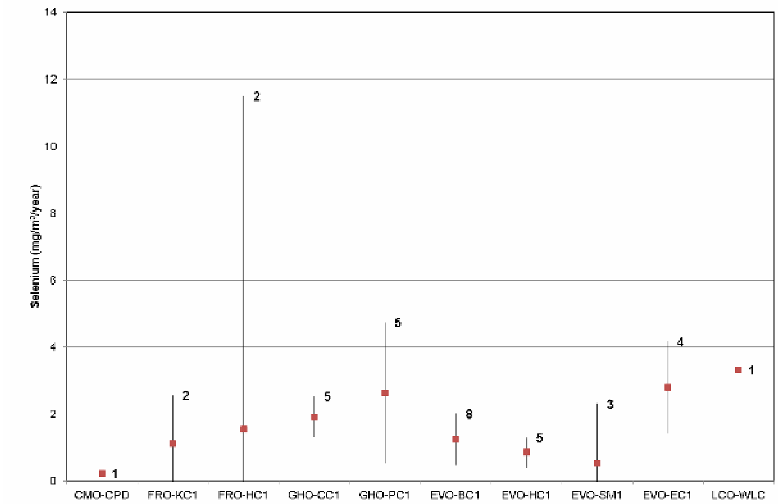


Figure 5. Average Annual Selenium Loads

Discussion

Direct investigation of geological materials from EVO (SRK 2004) and LCO (Kennedy et al. 2012) shows that selenium likely originates by oxidation of sulphide minerals (mainly pyrite) so that selenium and sulphate release by weathering ought to be correlated. The presence of a weak correlation between average annual selenium and sulphate loading from full scale waste rock dumps (Figure 6) appears to confirm the conceptual model and indicate that under the dominantly basic conditions selenium is released and transported with conservation of mass in solution. Due to the propensity for selenium in lower oxidation states (selenite, elemental selenium, selenide) to retard selenium leaching, it appears that selenium is dominantly present as selenate (SeO_4^{2-}). This appears to indicate that biological mechanisms capable of catalyzing reduction of selenium have limited effectiveness in these dominantly sub-aerial waste rock dumps.

Some differences in the ratio of selenium to sulphate loadings were apparent. The highest ratio is for LCO-WLC. There is no apparent reason for this difference which has persisted over 5 years of monitoring and is not unique to the single year when reliable flow data were available to calculate loadings. It implies either that Se is relatively more abundant at LCO compared to other sites or that release mechanisms for Se and SO_4 result in preferential release of Se compared to SO_4 at LCO.

The very distinctive chemistry for CMO-CPD may reflect the influence of different geological formations when compared to the other sites.

The apparent downward trend in Se/SO_4 ratios for sites others than CMO-CPD as overall concentrations decrease could reflect a number of factors. The most plausible explanation is that background waters have lower Se/SO_4 compared to waste rock contact waters and the decrease in ratio is simply a result of mixing of waters of different composition. Background waters tends to have very low Se (less than 1 µg/L) and SO_4 concentrations near 10 mg/L which is a ratio of less than 0.0001 mg Se/mg SO_4 compared to 0.001 0.0001 mg Se/mg SO_4 for waters with higher concentrations. A second possibility is that waters distant from sources may have been affected by processes which attenuate selenium. Attenuation of selenium in settling ponds has been documented in the Elk Valley (Martin et al. 2011) though these environments are not expected to result in significant sulphate reduction. This may be the case for EVO-SM1 which is monitored at the outfall of a settling pond 2.4 km from the waste rock dump. Age and waste rock dump geometry and location may be factors which could affect internal weathering and leaching conditions. For example, the waste rock dump above EVO-SM1 is also the smallest and least active in the dataset (Table 1) though it is also affected by significant background water dilution and the settling pond.

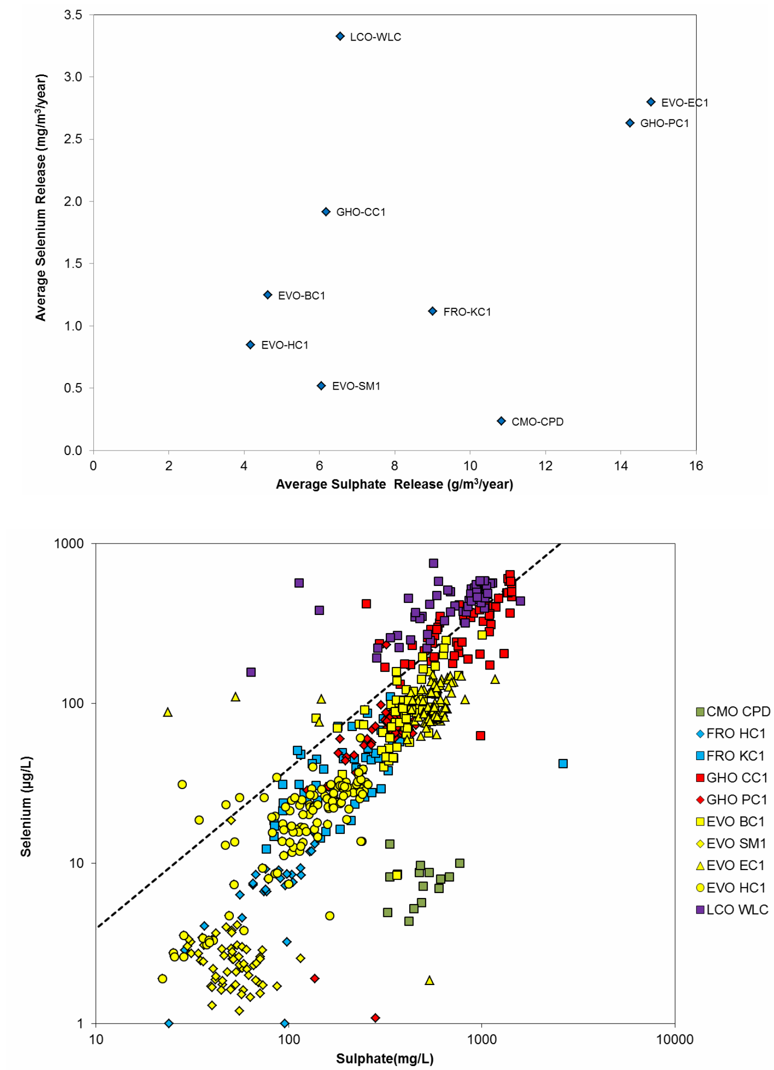


Figure 6. Comparison of Selenium and Sulphate Release (top) and Concentrations (bottom). The dashed line in the concentration graph is a constant Se/SO_4 ratio for reference.

An important finding from this interpretation of monitoring data is that both selenium and sulphate loads expressed in terms of waste rock volume vary over a narrow range despite the presence of a wide range of rock volumes and placement locations. For example, the difference between EVO-SM1 (inactive 9 million BCM placed near the headwaters of Six Mile Creek) and FRO-KC1 (active 970 million BCM at various locations in the Kilmarnock Creek catchment basin) is not significant when the range of uncertainty is considered. Two monitoring locations with somewhat similar placement locations (mainly headwater placement at GHO-PC1 and EVO-EC1) but with different waste volumes (79 and 365 million BCM) produced very similar sulphate and selenium loadings. The relatively small differences between observed loadings may reflect a number of factors in addition to the geochemical characteristics of the waste rock including uncertainty in the quantity of spoil, proportion of actual flow moving as groundwater rather than surface water and reliability of the flow measurement method.

Overall, this finding appears to indicate that either the entire waste rock volume or a consistent proportion of the waste is active in contributing sulphate and selenium loadings. This finding is also consistent with the valley wide observation that selenium concentrations in the Elk River have increased as total waste volumes have also increased (Figure 2). The strong correlation of seasonal flows with loadings which are the main source of annual loadings implies that once wetted the component of rock contributing to loadings responds rapidly to flow events resulting in little apparent lag between peak flows and loadings.

Conclusions

Selenium is released by oxidation of pyrite into dominantly basic drainage from coal mine waste rock dumps in the Elk Valley. The dominant aqueous selenium ion is inferred to be selenate based on consistent relationships between selenium and sulphate release which shows that selenium does not appear to be significantly in attenuated in the weathering environment.

The relatively narrow range in large-scale selenium and sulphate loading from the waste rock dumps spanning nearly three orders of magnitude in volume implies that sulphide oxidation rates are similar throughout the Elk Valley and that a consistent proportion of the rock mass is contributing to selenium loadings regardless of waste rock dump scale. Seasonal patterns of release of sulphate and selenium showing no detectable lag between high flow events and high loading release implies that selenium is leached from components of the waste rock dumps which transmit flows rapidly.

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Acronyms

BCM	Bank cubic metres
CCR	Coarse coal rejects
CMO	Coal Mountain Operations
EVO	Elkview Operations
EVSTF	Elk Valley Selenium Task Force
FRO	Fording River Operations
GHO	Greenhills Operations
LCO	Line Creek Operations
Se	Selenium
SP	Settling pond
TSS	Total suspended solids