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ISSN 0970-9274 ONLINE: ISSN 2456-6608
J Hum Ecol, 47(2): 155-162 (2014)
DOI: 10.31901/24566608.2014/47.2.06

Sand Mining Impacts on Heavy Metal Concentrations in Two Important River Systems of Northern Kwazulu-Natal, South Africa

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KEYWORDS Sand Mining. Heavy Metal. Mvoti. Geochemical Impacts

ABSTRACT Sand mining of the bed, banks, riparian zone and floodplains of rivers are known to cause major morphological and hydrological changes that impact both on their functioning and on riverine habitats and biota. In this study, the impacts of sand mining on sediment textural characteristics and heavy metal concentrations of twelve metals (Al, As, Cr, Cu, Fe, Pb, Mn, Ni, P, Ti, V and Zn) and three nutrients, Ca, Mg and P and their impacts for instream and riparian biota are presented for two important river systems, the Mvoti and the Mdloti Rivers of KwaZulu-Natal, South Africa. With the exception of Mn in the Mvoti system, the concentrations of all elements increased downstream of the sand mining operations. This is echoed by the Contamination Factor, Combined Pollution Index, and Pollution Load Index analyses. Enrichment Factor (EF) analysis indicates a slightly variable picture in that metals Pb and Ti for the Mvoti and Cu for the Mdloti as well as nutrients Ca and Mg for the Mvoti and Ca and P for the Mdloti decreased downstream of mining operations. Despite the variable EF, these results clearly highlight the importance of monitoring sediment geochemistry of sand mining operations, particularly for their potential hazardous impacts on downstream habitats.

INTRODUCTION

Sand mining of the riverine environments are known to cause major biological and morphological disruptions. The changes to channel geometry are multifarious and include depth and width modifications through the reduction and/ or elevation of the river bed, widening or narrowing of channels; the creation of artificial dams, bridges and access roads through the channel itself. These cascade into flow velocity changes; increases in turbidity and suspended sediment transport immediately downstream of the zone of activity; river discharge modification and therefore general alteration of the river's flow hydraulics.

In the past, Environmental scientists have focussed on studying these impacts and the downstream changes on the physical properties of water, such as increases in turbidity and suspended sediment levels (Ako et al. 2014; Barling and Moore 1994; Budd et al. 1987; Eyre and McConchie 1993; Kondolf and Swanson 1993; Walker 1999; Sukdeo et al. 2010; Govender 2009). Whilst the granulometric properties of mined sediment has historically been well researched, little attention has been accorded to the geochemical changes that occur particularly with regard to the downstream bed sediment although there have been some reporting of downstream water quality changes as a consequence of sand mining (for example, Sukdeo 2011). The textural character of downstream sediments is drastically altered due to sand extraction. Removal of coarser, sand-sized sediment grades leads to concentration of fine material on the river bed and banks downstream of the mining operations.

Fluvial sediments generally reflect the parent geology of the catchment from which they are derived (Schumann et al. 1999) and therefore their potential geochemical contribution to the environment. The textural characteristics of sediments are important in determining the extent to

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which they may serve as sinks for contaminants, particularly heavy elements whilst large grained particles are important in shaping the morphology of river and estuary systems (Villars and Delvigne 2001). Here, contaminants tend to adhere to fine grained sediment such as clays and silt that carry surface charges. Fine sediment deposits therefore have the potential to integrate contaminates over time while surface layers are in constant flux with the overlying water column (Binning and Baird 2001). Thus, coarse grained sediment plays less role in contaminant transport and their extraction through sand mining. This leads to potential pre-concentration of heavy metals on the remaining, predominantly fine grained sediment load downstream of mining operations.

Sediment-associated pollutants can influence the concentrations of metals in both the water column and biota if they are desorbed or become available (Milenkovic et al. 2005). In hydro-dynamically low energy environments (such as downstream abandoned pools following completion of sand excavation), where there is little sediment redistribution, contaminants can accumulate in bottom sediment to concentrations high enough to adversely affect organisms (Chapman 1992). The detection of elevated concentrations of contaminants such as heavy metals in sediment is a good indication of human-induced pollution and high levels of contaminants can often be attributed to anthropogenic influences, rather than natural enrichment of the sediment by geological weathering (Davies et al. 1991; Binning and Baird 2001).

The aim of this study was to assess the impacts of sand mining on two river catchments in KwaZulu-Natal, South Africa. The study will attempt to quantify downstream changes in sediment heavy metal and nutrient geochemistry and outline the need for pre-feasibility assessment of the physiographic and ecological viability for mining.

Regional Setting

The catchments of the Mvoti and Mdloti rivers are located in the province of KwaZulu-Natal (KZN) along the eastern coast of South Africa (Figs. 1a and b). The Mdloti is centrally located along the KZN coastline while the Mvoti is located approximately 50 km further north. The rivers flow through incised valleys (Pillay 1996), are dominantly perennial, and sediment yields are often high (Cooper 1993; Abed 2009). The coast of KwaZulu-Natal has a warm, humid climate with an average annual rainfall that exceeds 1000 mm (Harrison 2004). The Mvoti River drains a catchment with an area of approximately 2730 km^2 (Wepener 2007) and has a mean annual runoff of 375 million m^3 (Malherbe 2006). Land use within the catchment varies across a range of different activities from commercial forestry to sugarcane. The town of Stanger is located in the lower catchment. Govender (2009) estimated the Mdloti catchment area at 550 km^2, and a mean annual runoff of 125 million m^3 for the river. Sugarcane plantations dominate much of this catchment and the town of Verulam is located part of the catchment.

The specific study areas (Figs. 1a, b), were confined to the lower reaches of the river systems where sand mining operations are actively pursued. This encompasses the lower 10 km of the Mdloti and the lower 50 km of the Mvoti system. The geology of both catchments is dominated by sedimentary rock outcrops with some Archaen granites occurring in the hinterland region. The central parts are underlain by glacial till deposits and, mudstones and shales of the Ecca Group. This is overlain in some parts by dark grey shales, with Natal Group sandstones and conglomerates commonly present in the mid- to upper reaches of the catchment (DWAF 2007).

Areas of concentrated sand mining activity were identified from aerial photographs of both catchments and were used to locate sampling sites. For the Mvoti catchment, extensive sand mining occurs 4.93 km upstream of the river mouth and extends along the river for a distance of 11.4 km. Thereafter, sporadic operations occur for further 16 km. Sampling site R2 (upstream of mining) was located at Glenmill, 44.5 km from the mouth, and Site R1 (downstream of mining) located 4.73 km from the mouth. On the Mdloti River, mining is limited to two large operations located between 7 km and 4 km upstream from the mouth. Here sites numbered 4 (upstream of mining) and 5 (downstream of mining) were used. The mode of operation differed at the two catchments. Large volumes of sand were removed from the bed of the Mdloti system by two different companies creating two large pools of water (a rectangular pool 300 m long and 60 m wide and, an oval pool 250 m long and 120 m wide) and stock piling of material along the river. In

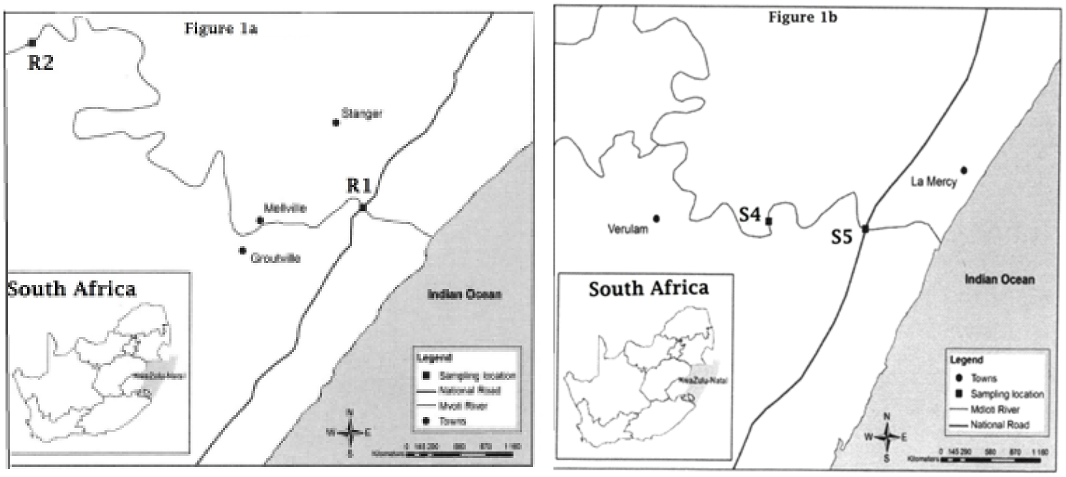

Fig. 1a and 1b. Map of the lower Mvoti River (1a) Map of the lower Mdloti River (1b)

the Mvoti system, numerous small to large scale companies strip away riparian vegetation and mine the bed, banks and floodplain areas creating extensive scarified patches.

MATERIAL AND METHODS

Fieldwork was carried out between March and May 2012. At each location, three samples were collected (one at midstream, and the others midway between both banks and midstream) using a Polyvinylchloride (PVC) Pipe Sediment Extractor, and stored on ice in polyethylene jars. Following collection, samples were analysed at the soil laboratories (granulometric assessment) and the analytical chemistry laboratory at the University of KwaZulu-Natal (Westville campus).

Textural analysis on sediment samples was carried out using a standard dry sieving technique (Abed 2009). Inductively-coupled plasma optic emission spectrometry (ICP-OES) was used to measure the concentrations of heavy metals aluminium (Al), arsenic (As), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), nickel (Ni), titanium (Ti), vanadium (V), and zinc (Zn) and, nutrients calcium (Ca), magnesium (Mg) and phosphorus (P) in the sediments. Inductively-coupled plasma techniques allow for several different elements to be simultaneously determined. The various elements emit both atomic and ionic spectra at the same time, by which they are identified.

A high metal concentration in sediment does not necessarily imply that contamination has taken place. These concentrations could signify the chemical composition and mineralogy of the parent rock. Hanson et al. (1993) stated that naturally occurring and anthropogenically introduced metals tend to accumulate in the same areas, highlighting why the mineralogical and granulometric factors that influence the natural variations of metal concentration in sediment should first be compensated for. This is usually done by geochemical normalization which normalizes metal concentrations to co-occurring conservative element (the normaliser or reference element) that provides the tracer of crustal decomposition (Kersten and Smedes 2002 as cited in CSIR 2010b). Iron was used as the normalizing element due to its abundance in the earth's crust (Rçsler and Lange 1972) resulting in its natural input into a system often far exceeding "unnatural inputs" (Harikumar and Jisha 2010). The Clarke Values for heavy metals were obtained from world averages for metals in sedimentary rock.

Contamination of sediments can then be geochemically expressed in a number of ways. Determination of the elemental enrichment factor (EF) is an important method used to assess anthropogenic contribution to contamination. The EF was calculated based on the abundance of the element present in the sample relative in the earth's crustal average concentration or background value (Harikumar and Jisha 2010) by the following equation (Martinez et al. 2007):

$$EF = \frac{[\text{Concentration Element}] / [\text{Concentration Fe}]}{[\text{Clarke Element}] / [\text{Clarke Fe}]} \qquad (1)$$

Where [Concentration Element] is the concentration of the element of interest; [Concentration Fe] is the concentration of Fe in the sediment; [Clarke Element] is Clarke value of the element for which enrichment is being determined, and [Clarke Fe] is Clarke value of Fe.

The EF was then used to differentiate heavy metals arising through natural or anthropogenic activities and was used to assess the degree of anthropogenic influence (Chakravarty and Patgiri 2009). As the value of EF increases, the contribution of anthropogenic influence also increases. The following categories are recognised on the basis of the EF (Sutherland 2000):

$EF < 1$ implies no enrichment; $1 \leq EF < 2$ implies minimal enrichment; moderate enrichment is indicated by $2 \leq EF < 5$; significant enrichment at $5 \leq EF < 20$; very high enrichment at $20 \leq EF < 40$ whilst $EF \geq 40$ implies extremely high enrichment.

The contamination factor (CF) was then employed to assess the relative contamination status of the sediment:

$$CF = \frac{[\text{Concentration of element in the sediment}]}{[\text{Background value of element}]} \qquad (2)$$

$CF < 1$ implies low contamination; $1 \leq CF < 3$ implies moderate contamination whilst $3 \leq CF < 6$ implies considerable contamination (Harikumar and Jisha 2010).

Assessment of the degree of contamination (Cd) defined as the sum of all contamination at a particular site (Håkanson 1980) was also calculated for each sample site. The following terminology is used to describe the contamination degree: $Cd < 6$ implies low contamination degree; $6 \leq Cd < 12$ implies moderate contamination degree whilst considerable contamination yields values of $12 \leq Cd < 24$. At $Cd \geq 24$ sediments are very highly contaminated.

The CF has been further developed as the Combined Pollution Index (CPI) by Abrahim and Parker (2008) which takes into consideration all possible contaminants assessed (n) 'at a site'. This index is therefore a cumulative value of the contaminant status and is given by:

$$CPI = (\Sigma\ CF)/n \qquad (3)$$

Classification of the CPI in terms of degree of contamination is as follows:

Very low: $CPI < 1.5$; Low: $1.5 \leq CPI \geq 2$; Moderate: $2 \leq CPI \geq 4$; High $4 \leq CPI \geq 8$; Very high: $8 \leq CPI \geq 16$; Ultra high: $CPI \geq 32$.

Finally, the pollution load index (PLI) developed by Thomilson et al. (1980) was applied to evaluate the extent of pollution at a site and is similar in principle to the CPI. It is determined by:

$$PLI = (CF1 \text{ X } CF\ 2 \text{ X...X } CFn)^{1/n} \qquad (4)$$

Where, CF refers to the contamination factor at each site and n refers to the number of elements for which the PLI is assessed.

RESULTS AND DISCUSSION

Granulometric Sediment Characteristics

Walker (1999) notes that much of lithologies from which the sediment of the Mdloti River derived have been considerably weathered, and that the depth of weathering exceeded 20 m in some parts of the catchment. Since a large portion of the catchment hinterlands of both river systems is underlain by Natal Group sandstones and partly by Natal Metamorphic Province granites and gneisses, the resultant sediment is a mixture of coarse to fine sand (± 70 %) and fine silt and clays (±30%) (Walker1999). Comparing sediment textures before and after the mining operations for both rivers reveals that a higher percentage of finer grades of sand occurs in the Mdloti system reflecting the greater degree to which sediment is weathered chemically as noted by Walker (1999) whilst a large fraction of the sediment in the Mvoti system lies in the silt size range likely reflecting the extent of mechanical micronizing in a relatively larger fluvial system (Table 1).

Texture triangular diagrams plots of sand sized sediment form six sites in each of the two rivers (Figs. 2a and 2b) displays a bi-modal distribution pattern for the Mdloti uniformly with very little medium grained sand whilst the Mvoti sediment shows more varied sediment distribution.

The importance of the textural differences lies in the target grain size mined at these systems. Mining operations target fine grained sand extraction leading to the concentration of the other grades immediately downstream of the operations as stockpiled 'waste' is returned to the fluvial system. An important consideration for this study is the degree to which the fine sediment composition, particularly clays, increases downstream. It is well documented that charged metal ions are readily adsorbed onto

Table 1: The mean textural composition of the upstream and downstream sediment for both rivers from ten sampling sites instances

Grade	*Size (mm)*	*Mdloti S4*	*Mdloti S5*	*Mvoti R2*	*Mvoti R1*
Coarse Sand	0.50–1	1.26	7.85	0.02	15.24
Medium sand	0.25–0.50	5.52	16.69	0.88	9.10
Fine sand	0.125–0.25	72.61	29.15	52.35	21.98
Very fine sand	0.0625–0.125	19.97	19.99	1.46	1.35
Silt	0.0039– 0.0625	0.53	14.02	44.78	51.02
Clay	< 0.0039	0.071	11.31	0.51	2.62

the electrically charged clay mineral surfaces (Callow 1994; Villars and Delvigne 2001). The settling of such water borne clays creates potentially metal enriched sediment layers. Re-suspension and desorption of these metals can lead to contamination of the aquatic environment and have detrimental effects on habitats. Table 1 shows that a considerable amount of clays accumulate downstream of the Mdloti mining operations (11.31 %) as compared to the Mvoti (2.62 %). The larger percentage gain in the Mdloti River is likely a consequence of the mode of mining operations where the river bed and banks have been extensively excavated (wet pit mining) creating stilling conditions allowing for the gradual settling out of fines. In the Mvoti system, although mining of the bed does occur, no extensive in-stream depressions have been created. Here, extensive mining of the riparian and floodplain zones (dry pit) characterize the operations.

Geochemical analysis

The mean concentrations of heavy metals, Enrichment Factors (EF) and Contamination Factors (CF) determined at each site are illustrated in the tables below in Tables 2 and 3.

Cursory examination of the comparative upstream and downstream metal concentrations for both rivers suggest fairly conclusive downstream increases in metal and nutrients as a consequence of the mining operations. The only exception to this trend was the downstream decrease in concentration recorded for Mn at site R2 on the Mvoti River. Other exceptions are for undetected elements (as in the Mvoti River and Ti in the Mdloti River). However, despite this obvious downstream trend, it is important to contextualize the relative increase in terms of the impacts of the sand mining operations on contaminant status using known geochemical indices. Accordingly, Tables 2 and 3 also con-

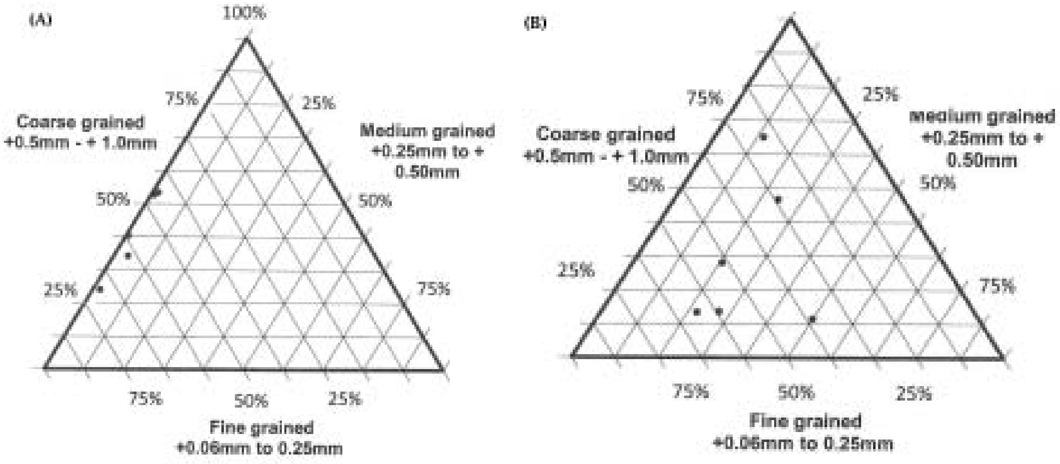

Fig. 2. Ternary diagrams showing sediment composition by grain size for sites along the Mvoti River (A) and Mdloti River (B) KwaZulu-Natal

Table 2: Chemical composition and geochemical index values of Mvoti River sediments taken upstream (R2-U) and downstream (R1-D) of sand mining activity

	Clark Value	*R2: (U)*	*R1:(D)*	*EF:R2(U)*	*EF: R1(D)*	*CF:R2(U)*	*CF:R1(D)*
Al	81300	67.95	365.7	0.355	0.373	8.358	44.982
As	5	0	0	0	0	0	0
Ca	36300	16.62	50.54	0.194	0.115	4.579	13.923
Cr	200	0.17	1.27	0.361	0.526	8.500	63.500
Cu	70	0	0.21	0	0.249	0	30.000
Fe	50000	117.8	603.6	1	1	23.560	120.720
Pb	16	0.05	0.23	1.326	1.191	31.250	143.750
Mg	20900	11.8	54.24	0.240	0.215	5.646	25.952
Mn	1000	10.45	7.6	4.435	0.630	104.50	76.000
Ni	80	0	0.14	0	0.145	0	17.500
P	1180	8.97	58.62	3.227	4.115	76.017	496.780
Ti	4400	10.17	48.48	0.981	0.913	23.114	110.182
V	150	0.26	1.91	0.736	1.055	17.333	127.333
Zn	132	0.03	0.65	0.096	0.408	2.273	49.242

Table 3: Chemical composition and geochemical index values of Mdloti River sediments taken upstream (S4-U) and downstream (S5-D) of sand mining activity

	Clark Value	*R2: (U)*	*R1:(D)*	*EF:R2(U)*	*EF: R1(D)*	*CF:R2(U)*	*CF:R1(D)*
Al	81300	18.34	71.04	0.503	0.847	2.256	8.738
As	5	0	0.03	0	5.813	0	60
Ca	36300	6.73	18.98	0.414	0.507	1.854	5.229
Cr	200	0	0.11	0	0.533	0	5.5
Cu	70	0.07	0.15	2.231	2.076	10	21.429
Fe	50000	22.41	51.61	1	1	4.482	10.322
Pb	16	0.06	0.18	8.367	10.899	37.5	112.5
Mg	20900	3.21	11.49	0.343	0.533	1.536	5.498
Mn	1000	0.53	2.08	1.183	2.015	5.3	20.8
Ni	80	0	0.01	0	0.121	0	1.25
P	1180	3.09	3.88	5.843	3.186	26.186	32.881
Ti	4400	-	-	-	-	-	-
V	150	0.11	0.28	1.636	1.808	7.333	18.667
Zn	132	0	0.1	0	0.734	0	7.576

tain the enrichment factors and contamination factors for each element.

Contamination factors (CF) analysis confirms that the downstream increase in all elements at both river sites demonstrate increases in contamination with the same exceptions noted above. In the Mvoti system, however, for most of the elements, the CF indicates more than a five-fold increase in contaminant status from upstream to downstream sites. This has caused the sediment downstream of the mining operations to become considerably contaminated.

In the Mdloti system, levels of contamination do not reach the same levels as the Mvoti system but is still a concern as most elements are categorised as moderate to considerably contaminated. For the Mvoti system, as a consequence of mining, increased enrichment over naturally expected concentrations had occurred for Al, Cr, Cu, Ni, P, V, and Zn of the Mvoti system but not for Ca, Pb, Mg and Ti. In the Mdloti, more elements displayed greater downstream increases in enrichment with only Ca, Cu and P recording decreases.

The Combined Pollution Index (CPI) for the Mvoti River is 0.003 upstream at R2 and 0.01 downstream of mining at R1. For the Mdloti River the upstream value was 0.001 as compared to downstream value 0.002. Whilst both indexes indicate a very low degree of contamination, the downstream increases are as consequences of the mining operations. The pollution load index (PLi), like the PCI, is an important assessment tool because it considers the accumulated pollution effect of all potential contaminants at a site. In this study, only two sites were used at each of the rivers (Fig. 3).

The CPI analysis and the CLi (Fig.3) clearly show that greater pollution loading occurs downstream at the Mvoti. The variation in contamina-

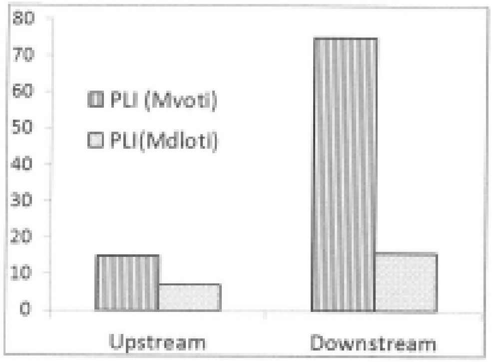

Fig. 3. Pollution Load indices (PLi) for upstream and downstream sampling sites of the Mvoti and Mdloti Rivers

tion of these rivers is due to a number of factors. The Mvoti River and catchment is significantly larger than the Mdloti. Hence Mvoti drainage covers a larger area and caries larger discharge and sediment load. Consequently, the potential for carrying a greater suspended and solute load is increased. Mining on the Mvoti occurs on a large scale, extending for a linear distance of 27.4 km along the river. Mining on the Mdloti occurs over only 3 km. Hence the greater disruption of in-stream sediments along the Mvoti River creates the potential for greater desorption of ions back into solution.

CONCLUSION

It is noted that the processes of prospecting, extracting, concentrating, refining and transporting of minerals have great potential for disrupting the natural environment. Whilst the mining of sand and gravel could not reach up to the status of other, more recognised minerals, it nevertheless, impacts on the environment in a multiple of ways that have negative consequences for a variety of aquatic, riparian and floodplain biota. This study focused on examining the changes in sand mining by a comparative study of sediment geochemistry upstream and downstream of mining operations along the Mdloti and Mvoti Rivers of KwaZulu-Natal, South Africa. Geochemical indices were used to make the comparison, which was confirmed with a general increase in metal concentrations downstream of mining operations with some minor variations. The type of mining operation, together with the granulometric character of upstream sediments was found to be important in downstream metal accumulation in sediments. Of the three nutrients studied, downstream decreases for Mg (Mvoti River) and P (Mdloti River) and, Ca (both rivers). It was evident that, sand mining operations do affect the downstream geochemistry of sediments with potential adverse impacts for biota. This study highlights the need for prior assessment of mining impacts, sound management and monitoring of operations as well as post mining restoration of the mined areas.

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