Seasonal Geochemical Variation of Sediments in the Sabie River, Mpumalanga, South Africa

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Abstract

Water pollution resulting from mining activities has become a major problem in South Africa. In this study, the seasonal variation of geochemical properties of solid wastes and stream sediments in the Sabie River catchment were characterised. There was no significant variation observed in terms of metal (loid) dispersion, namely, Cr, Zn, Cu, Ni, Pb and As during wet and dry seasons respectively. Furthermore, the mineralogy of sediments indicated that acid-producing minerals such as hematite and jarosite, typically found in mine wastes are potential sources of pollution to the Sabie River. In addition, acid-neutralizing mineral dolomite found in the sediments can act as buffers for potential acid. There was no evidence of metal dispersion from the Nestor tailings storage facility to the adjacent water resources, Klein-Sabie and Sabie River respectively.

Keywords: Stream Sediment, Sabie River, Acid-Producing Minerals, Acid-Neutralizing Minerals, Toxicity

Introduction

Together with the Komati and Crocodile basins, the Sabie River basin is part of the Inkomati River basin. Originating at an elevation of 2, 207 m.a.s.l., and the river flows in an easterly direction and passes through the Kruger National Park (KNP) towards the confluence with the Inkomati River in Mozambique. The river flows through commercial forestry plantations (pine trees and eucalyptus), sawmills, trout farms, fishery areas, waste treatment works, defunct mines of the Transvaal Gold Mining Estate (TGME), industrial and agricultural activities which are dominant in its upper reaches. Also located in the upper reaches is the town of Sabie while in the lower reaches of the basin there is the Hazyview town, rural settlement and Kruger National Park (KNP).

Recent studies carried out in the Sabie-Pilgrim's Rest goldfields showed a necessity of studying the geochemical composition of stream sediment along the Sabie River catchment (Novhe *et al.*, 2014) (Rudzani *et al.*, 2017); (Rudzani *et al.*, 2018); (Rudzani, Novhe and Mashalane, 2019). Based on the findings, concentrations of metals such as As, Cr, Cu, Pb, Ni and Zn were elevated on mine wastes. Furthermore, the Nestor tailings storage facility (TSF) had no vegetation cover and susceptible to water and wind erosion which may consequently enrich the surrounding environment with toxic metals. The main aims of this study were to determine the concentration of major, trace and rare earths elements and define anthropogenic versus geogenic distribution in the Sabie River catchment with seasons. The data on the distribution of these metals in sediment near the mine wastes and along the catchment could provide valuable information on risk and exposure assessment of communities near the mine site.

Study Area

The Sabie River system is one of the largest rivers in the Mpumalanga Province, situated within the Inkomati water Management Area and originates at an elevation of 2207 m.a.s.l., and enters the Kruger National Park (KNP) 81.3 km downstream from its source (Figure 1).

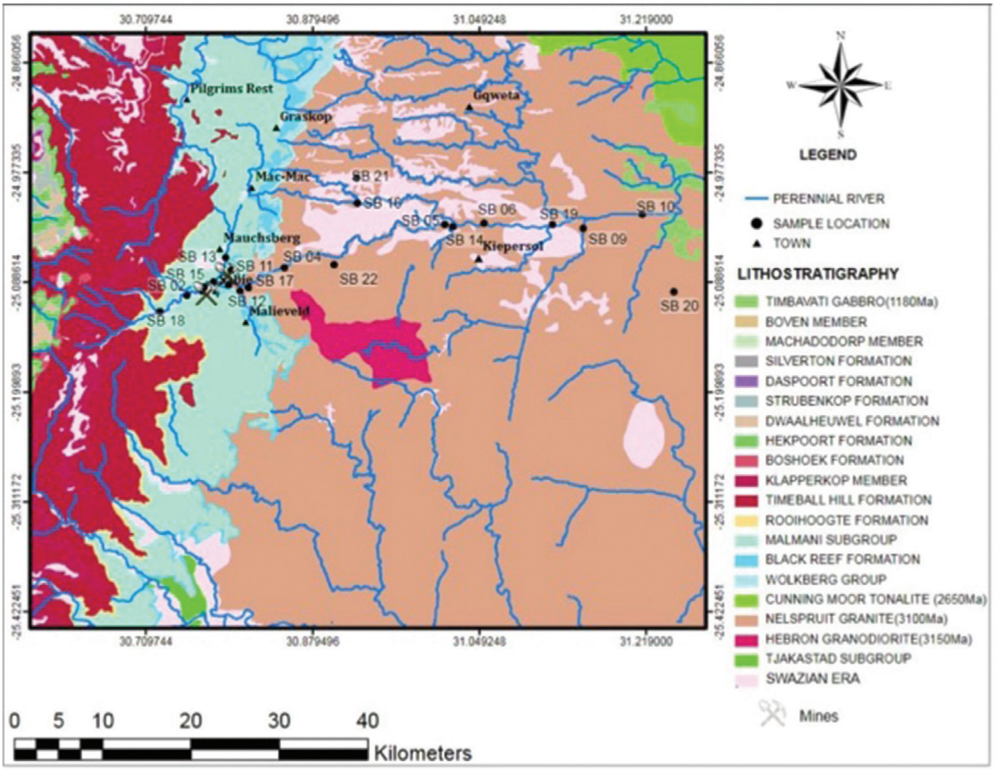

Figure 1 *Location of the Sabie River catchment and sample sites for collection in this project (CGS 1:250 000).*

Geologically, the area comprises of rocks of the Nelspruit Suite ranging from the Swazian era (Nelspruit granite and Hebron granodiorite) and 2800 Ma Cunning Moor Tonalite, Wolkberg Group, Black Reef Formation, Chuniespoort Group (Malmani Subgroup) and Pretoria Group (Rooihoogte, Timeball Hill, Boshoek, Hekpoort, Dwaalheuwel and Strubenkop Formations; Figure 1).

Materials and methods

Sampling and laboratory analyses

Sediments eroded from the Nestor TSF were collected and analysed for the chemical and mineralogical compositions (SB00) and were compared with twenty (20) composite stream sediment samples that were randomly collected to a depth of 15 cm (SB1-21; Figure 1) from Sabie River catchment. Samples were collected at various distances from agricultural areas, industrial areas, mining areas and urban/commercial areas. Sampling was done during November and December 2019, representing a wet season as well as July and August 2020 (dry season). Stream sediment samples were collected in the Sabie basin along the acid mine drainage (upstream, in the middle and downstream) in order to assess their impact on the dispersion of metals on the receiving water. The sediment samples were collected using a shovel with a narrow blade, bucket for homogenization of sediment samples, and plastic bags that were tightened to avoid oxygen penetration and to preserve biological or chemical equilibria. The samples were analysed by X-ray florescence (XRF) spectrometry and X-ray diffraction (XRD) spectrometry.

Data analysis

Assessment of metal pollution in sediment

Results and Discussion

The degree of contamination from the metal species in the Sabie River catchment was evaluated by determining the contamination factor (CF), pollution load index (PLI) and geoaccumulation index (*Igeo*).

Enrichment factor (EF)
The enrichment factor (EF) is an index/indicator used for reflecting the level of environmental contamination. In this study, EF was employed to evaluate possible anthropogenic input of metal species to the Sabie River catchment and was calculated as:

$$EF = (\frac{Cn}{LV})\ sample/(\frac{BB}{LV})\ background$$

where [Cn/LV] sample is the concentration of analysed metal and one of the following metals Fe/Al/Ca/Sc/Ti/Rb and [GB/LV] *background*-reference concentration of analysed metal (Cn) and one of the following elements Fe/Al/Ca/Sc/Ti/Rb (LV). For this study Rb was used as a reference element and SB18 (Mac Mac River) was selected as a background as there are no mining activities and little anthropogenic input. Islam *et al.* (2015c) proposed the ratio of measured concentration to natural abundance as a contamination factor (CF) and classified into four grades for pollution monitoring of a single metal over a period of time: low degree ($CF<1$), moderate degree ($1 \leq CF<3$), considerable degree ($3 \leq CF<6$), and very high degree ($CF \geq 6$).

Geoaccumulation index (I*geo*)
In order to evaluate the degree of metal pollution of sediment by comparison to the baseline metal concentration of the surrounding area, the geoaccumulation index was used. By definition, the geoaccumulation index is:

$$Geoaccumulation\ index\ (Igeo) = log2\ (\frac{Cn}{1.5Bn})$$

where C*n* is the toxic element concentration in a sample and B*n* is the measured concentration of the element in unpolluted sediments. Other studies had used similar approach to characterize pollution of sediments including (Sarmiento *et al*., 2011); (Han *et al*., 2017). Samples were classified as unpolluted (I*geo* < 1) and highly polluted (I*geo* > 5).

Chemical index of alteration (CIA)
The chemical index of alteration (CIA) was used to evaluate the extent of weathering of feldspars relative to unaltered rocks from the Sabie catchment area. Nesbit and Young (1982) defined as:

$$CIA = (Al_2O_3/Al_2O_3+CaO^*+Na_2O+K_2O) \times 100$$

where CaO* is the amount of CaO incorporated in the silicate fraction of rocks. According to Taylor and McLennan (1995), the average upper continental crust denoted a CIA value of 47. This implies that CIA values of between 45 and 55 indicate weak weathering conditions and a value of 100 represent extreme weathering which is supported by the presence of typical weathering minerals such as kaolinite and gibbsite.

Two sediment quality guidelines (SQGs) for metals in freshwater ecosystems described by McDonald *et al.* (2000) were used to describe the possible toxicity levels of metal species in the Sabie River catchment. In SQGs, samples were grouped into two categories: threshold effect levels (TELs) and probable effect concentrations (PECs). The TELs are the concentrations below which harmful effects are unlikely to be observed and the PECs are the concentrations above which harmful effects are likely to be observed (McDonald *et al*., 2000).

Statistical analysis
The Pearson R correlation analyses were applied to assess elemental associations and origins of analysed elements using software Stata version 13. Critical values of the correlation coefficient (r) 0.81 at $p \leq 0.05$ were considered highly significant.

Geochemistry of solid wastes and stream sediment

Major elements in mine waste and stream sediments
The results of the XRF analysis of the major elements presented as percentages of the corresponding oxides. The following oxides were observed to be dominant in all sampling sites assessed: SiO_2 (40.24-86.15%), Al_2O_3 (5.67-52.20%), Fe_2O_3 (1.62-11.75%), K_2O (1.02-4.45%), Na_2O (0.08-2.87%), CaO (0.12-1.87), Ti_2O (0.23-1.69%), MgO (0.30-1.63%), MnO (0.018-0.273%), P_2O_5 (0.030-0.270%) and Cr_2O_5 (0.007-0.102%). More than 75% of major elements in all surficial samples were accounted

for Si-Al-Fe component, which is likely reflecting a relatively high quartz, feldspar, clay minerals and mica present in the sediments. The dominant elements such as Si, Al, Fe, Ti and Mn can be potentially toxic to aquatic environment.

According to (Nesbitt and Young, 1982) CIA values of between 45 to 55 indicate that the rocks of the catchment area undergo moderate degree of weathering and value of 100 indicates intense weathering. CIA value for Nestor TSF was 74.04 and varied from 53.19 to 88.58 in wet and dry seasons respectively in the Sabie River (SB01-09). The high CIA values were also observed at Lone Creek (SB10) at 88.46 and 82.33 in wet and dry seasons respectively. This implies that there is wide range of chemical weathering occurring in the Sabie River. However, low CIA values were recorded at sites SB08, SB16, SB17, SB18, SB20 and SB 21, which are underlain by granitic rocks.

Pearson correlation analysis (Table 1) showed the significant positive correlation Ti with both Fe and Al, Cr and Mg, as well as P and Fe. The zero to highly significant correlation between Si and other elements (Ti, Al, Fe, Mn, Mg, Ca K, P, Na, and Cr) suggests their removal from silicate phase during weathering. The highly positive correlation of Al with Ti and Fe, Ca with Mg and Cr with Mg and Ca indicate similar input sources and/or very close mineral association between these groups of elements.

Trace elements concentrations in mine waste and stream sediments

Trace element concentrations (As, Co, Cu, Cr, Ni, Pb, V and Zn) of mine waste (SB00) and surficial sediments (SB01-21) and their mean, minimum, maximum and standard deviation values in the Sabie River catchment gathered in Table 2 were used to evaluate metal their distribution. In addition, the mean upper crust concentrations (UCC- Rudnick and Gao, 2003) and the toxic effect concentrations and probable effect concentrations (MacDonald *et al.*, 2000) for freshwater sediments for a selection of trace elements is shown. Analysis revealed that the mean sediment trace element concentration increases in the following order: Pb<Co<Ni<Cu<Zn<As<Cr. Metal species concentrations in mine waste (SB00) were higher than upper crustal crust concentrations (Rudnick and Gao, 2003). However, metal species in mine waste sample are lower than in the stream sediments. The mean concentration of As in sediment was observed 92.7 mg/kg in summer and 80.4 mg/kg in winter respectively which were higher than average upper continental crust. The chromium concentration in the sediment was higher than other metals because of direct discharging untreated wastes from fertilizers (SB01-21). Higher values of Cr recorded during summer season at SB21 (Bega River) indicates its input, which might be originated from residential and industrial wastes.

The concentrations of As, Cr, Cu and Ni in some sediment samples exceeded the consensus-based PEC for freshwater ecosystems, implying probable adverse effect on the ecosystem (Table 2). All mean

***Table 1** Pearson correlation coefficient between major elements.*

	SiO_2	TiO_2	Al_2O_3	Fe_2O_3 (t)	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Cr_2O_3
SiO_2	1.00										
TiO_2	-0.93	1.00									
Al_2O_3	-0.90	0.83	1.00								
Fe_2O_3 (t)	-0.87	0.91	0.62	1.00							
MnO	-0.24	0.32	0.07	0.46	1.00						
MgO	-0.47	0.42	0.45	0.41	0.14	1.00					
CaO	-0.17	0.06	0.20	-0.03	-0.08	0.79	1.00				
Na_2O	0.22	-0.33	0.09	-0.60	-0.45	-0.09	0.38	1.00			
K_2O	0.09	-0.20	0.26	-0.46	-0.40	-0.18	0.02	0.75	1.00		
P_2O_5	-0.55	0.45	0.25	0.60	0.16	0.08	-0.04	-0.39	-0.32	1.00	
Cr_2O_3	-0.33	0.31	0.17	0.40	0.38	0.77	0.62	-0.27	-0.47	0.17	1.00

values recorded for As were much higher than consensus based TECs and PECs. This implies a probable adverse effect due to the As-presence in the Sabie River system. A consistent result is also yielded from the calculated geoaccumulation index of As which reached 5.90 and 8.73 in wet and dry seasons respectively (Table 4). There is also probable a Cr and Ni pollution potential in the Bega River (SB21) based on the geoaccumulation index while drainage next to the old Rietfontein Mine (SB15) was observed as a potential Cu-Pb pollutant to the surrounding ecosystem. In addition, geoaccumulation indexes of elements such as Co, Cr, Cu, Pb and Zn showed classes unpolluted to moderately polluted at most sites. This implies that the pollution caused by mine wastes regarding toxic elements may be likely limited to As.

Metal concentrations were high in winter than in summer because of low water flow in winter that can assist toxic metals species to accumulate in sediments.

Pearson correlation matrix showed the following significantly positive correlations during the dry season (shaded): Ni with Co and Cr, Pb with Co and Ni, Zn with Ni and Pb. In wet season, the following correlation was observed (unshaded): Co with Ni and Zn, Cr with Ni, Cu with Pb and Zn with Ni and Pb (Table 3).

The geoaccumulation index of sediments showed that sediment are extremely

Table 2 *Trace elements concentrations and their mean, maximum and standard deviation in the Sabie River catchment compared to threshold effect concentration and probable effect concentration (PEC) (MacDonald* et al., *2000).*

Sample number	As (ppm)		Co (ppm)		Cr (ppm)		Cu (ppm)		Ni (ppm)		Pb (ppm)		Zn (ppm)	
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
SB00	365	_	11	_	135	_	77	_	32	_	38	_	27	_
SB01	23	17	37	38	263	277	86	78	101	103	23	21	89	90
SB02	7.9	15	19	36	214	272	32	65	37	94	17	31	49	73
SB03	382	293	38	36	192	204	79	75	77	82	18	18	135	109
SB04	56	322	15	35	120	155	31	222	25	73	12	24	31	96
SB05	271	277	25	31	110	136	103	159	49	65	16	20	76	93
SB06	186	279	19	17	108	111	52	41	38	38	13	13	55	47
SB07	149	Bdl	17	14	93	89	40	20	33	36	11	16	43	58
SB08	32	13	11	Bdl	69	78	20	6.3	18	7.8	12	5.5	36	9.3
SB09	17	16	7.3	62	81	241	10	68	17	101	7.1	14	15	64
SB10	16	19	33	33	236	268	73	63	83	92	18	20	83	83
SB11	23	18	22	19	253	382	35	27	41	38	27	13	45	25
SB12	170	234	34	43	183	202	67	176	56	107	12	39	85	294
SB13	13	11	61	3.2	211	52	58	7.9	85	15	11	8	55	7.1
SB14	133	186	74	89	162	207	111	147	84	87	27	29	64	49
SB15	90	81	13	18	19	24	420	1180	13	16	52	25	36	41
SB16	Bdl	Bdl	6.1	Bdl	39	24	7.0	4.5	14	10	9.5	9	24	14
SB17	Bdl	Bdl	4.4	11	60	120	6.4	15	18	47	7.7	12	11	39
SB18	13	7.4	21	25	59	81	31	33	28	42	7.5	6.7	26	17
SB19	Bdl	Bdl	6.4	Bdl	38	23	8.4	4.3	9.4	6	4.6	3.2	13	5.4
SB20	Bdl	89	5.1	9.8	35	85	6.8	19	8.8	22	9.2	8.9	14	23
SB21	Bdl	Bdl	38	10	710	240	40	9.7	172	41	16	8.7	80	25
Mean	92.7	80.4	24.3	25.2	159.5	155.8	66.1	115.3	48.8	53.5	17.1	16.4	50.8	80.6
SD	120.5	118.7	18.2	22.0	146.9	100.3	86.9							
UCC	4.8		26.0		92.0		28.0		47.0		17.0		67.0	
TEL	9.79				43.4		31.6		22.7		35.8		121	
PEC	33.0				111		149		48.6		149		459	

Bdl, below detection limit; SD, standard deviation; TEC, toxic effect concentration; PEC, probable effect concentrations UCC: upper continental crust (Rudnick and Gao, 2003).

***Table 3** Pearson correlation coefficient between trace elements in the Sabie River.*

	As	Co	Cr	Cu	Ni	Pb	Zn
As	1	0.15	-0.08	0.24	0.05	0.34	0.47
Co	0.41	1	0.48	0.14	0.73	0.16	0.64
Cr	0.08	0.56	1	0.48	0.91	0.08	0.53
Cu	0.19	0.12	-0.18	1	0.02	0.82	0.02
Ni	0.37	0.83	0.74	-0.04	1	0.1	0.71
Pb	0.50	0.69	0.48	0.40	0.76	1	0.17
Zn	0.54	0.47	0.37	0.12	0.71	0.79	1

contaminated with As at some sites, especially in winter (dry season) (Table 4).

The calculated enrichment factor values for the Sabie River catchment for trace elements revealed minor to severe enrichment in sediments (Table 5). The values of contamination factor (CF) Co showed moderate contamination (1≤CF<3), whereas all other trace elements (As, Cr, Cu, Ni<Pb and Zn) showed the degree of contamination (CF>6), though values for Cr, Cu, Ni and Pb varied with seasons and were lower than six depending on seasonal change.

Mineralogy of solid wastes and stream sediment

The exposed and underlying lithologies of the Sabie River catchment are dominantly composed of gneiss, granite, quartzite and dolomite. Therefore, surficial sediment samples of mineralogy of the streams closely related to the prevailing rock outcrops. There is great variation in terms of mineralogy of the Nestor TSF sediments and sediments collected from the streams. The mineralogical assemblage of the stream sediment is mostly comprised of quartz (51-87%), followed by feldspars (K-feldspar (1-21%), while phyllosilicate (mica and clay minerals) have concentrations varying from 0 to 15%, and were absent on tailing sediments (Table 6). In addition, dominant on sediments samples is the secondary mineral gibbsite especially on the upper reaches of Sabie River. This is supported by the presence of clay mineral kaolinite. Carbonate mineral dolomite occurred in few locations in the upper part of the catchment at very low concentrations of 0 to 6% (SB02) and near Glynns Lydenburg TSF (SB11). In addition to the minerals detected from stream sediments, the TSF sample contained two acid producing minerals primary mineral hematite (2 wt.%) and secondary mineral jarosite (4 wt.%). Traces of smectite were only found sporadically in the catchment area.

There was significant seasonal variation observed in the predominant pattern of sediment mineralogical composition throughout the catchment area.

Conclusions

There was significant variation in most trace metal concentrations as they were high in winter than summer season. Low concentrations of metal species occurred in

***Table 4** Geoaccumulation index of trace metals in sediment of Sabie River catchment.*

	Igeo (As)		Igeo (Co)		Igeo (Cr)		Igeo (Cu)		Igeo (Ni)		Igeo (Pb)		Igeo (Zn)	
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
Min	0.00	0.00	0.04	0	0.04	0.05	0.04	0.00	0.09	0.04	0.12	0.10	0.10	0.08
Max	5.64	8.73	0.71	0.71	2.40	0.90	2.70	0.71	1.23	0.54	1.38	0.93	1.04	1.28

***Table 5** Contamination factor of trace metals in the Sabie River catchment.*

	CF (As)		CF (Co)		CF (Cr)		CF (Cu)		CF (Ni)		CF (Pb)		CF (Zn)	
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
Min	0.00	0.00	0.17	0.00	0.09	0.52	0.18	0.08	0.13	0.08	0.90	0.76	0.48	0.34
Max	49.62	48.19	3.05	3.02	12.00	4.93	4.26	7.45	6.19	3.27	5.56	7.22	8.60	22.07

Table 6 *Mineralogical composition of tailings and stream sediments in the Sabie River catchment.*

Sample number	Dolomite		Hematite		K-Feldspar		Quartz		Mica		Kaolinite		Gibbsite		Smectite	
	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry
SB00	Bdl	_	2	_	Bdl	_	85	_	9	_	Bdl	_	Bdl	_	Bdl	_
SB01	Bdl	Bdl	Bdl	Bdl	1	1	71	57	7	15	5	15	5	4	6	Bdl
SB02	Bdl	6	Bdl	2	14	1	53	60	5	11	1	11	Bdl	2	Bdl	Bdl
SB03	Bdl	Bdl	Bdl	1	2	9	88	69	4	8	2	8	2	3	Bdl	Bdl
SB04	Bdl	Bdl	Bdl	2	16	17	64	62	3	7	2	7	3	4	Bdl	Bdl
SB05	Bdl	Bdl	Bdl	1	10	5	72	69	3	7	3	11	3	5	Bdl	Bdl
SB06	Bdl	Bdl	Bdl	1	21	7	51	76	3	2	2	3	2	1	Bdl	Bdl
SB07	Bdl	Bdl	Bdl	Bdl	14	9	61	72	Bdl	3	Bdl	3	Bdl	Bdl	Bdl	Bdl
SB08	Bdl	Bdl	Bdl	Bdl	14	7	67	87	Bdl	1	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
SB09	Bdl	Bdl	Bdl	1	10	1	73	85	1	4	Bdl	2	Bdl	5	Bdl	Bdl
SB10	Bdl	Bdl	Bdl	1	2	Bdl	58	52	11	20	4	17	10	6	4	Bdl
SB11	3	8	Bdl	Bdl	4	4	86	74	Bdl	4	Bdl	3	Bdl	1	Bdl	Bdl
SB12	Bdl	Bdl	Bdl	1	8	1	84	84	4	5	Bdl	4	2	3	Bdl	Bdl
SB13	Bdl	Bdl	Bdl	Bdl	Bdl	21	88	67	Bdl	1	Bdl	1	12	Bdl	Bdl	Bdl
SB14	Bdl	Bdl	Bdl	Bdl	7	4	93	85	Bdl	7	Bdl	3	Bdl	Bdl	Bdl	Bdl
SB15	Bdl	Bdl	Bdl	1	34	21	22	55	10	6	17	4	Bdl	Bdl	Bdl	Bdl
SB16	Bdl	Bdl	Bdl	Bdl	14	11	61	80	Bdl	1	Bdl	1	Bdl	Bdl	4	Bdl
SB17	Bdl	Bdl	Bdl	Bdl	17	7	71	62	Bdl	2	Bdl	5	Bdl	Bdl	Bdl	Bdl
SB18	Bdl	Bdl	Bdl	Bdl	10	6	72	84	1	2	Bdl	1	2	3	Bdl	Bdl
SB19	Bdl	Bdl	Bdl	Bdl	4	3	85	94	Bdl	1	Bdl	1	3	1	Bdl	Bdl
SB20	Bdl	Bdl	Bdl	1	14	17	55	71	1	3	2	2	Bdl	1	Bdl	Bdl
SB21	Bdl	Bdl	Bdl	Bdl	17	7	52	73	9	1	Bdl	Bdl	Bdl	Bdl	Bdl	2
Mean	0.1	0.6	0.1	0.6	10.6	7.6	68.7	72.3	3.2	5.3	1.7	4.9	2.0	1.9	0.6	0.1
SD	0.6	2.1	0.4	0.7	8.6	6.5	16.8	11.7	11.7	5.0	3.7	4.9	3.3	2.0	1.7	0.4

stream sediments compared to mine wastes. The contamination factor, pollution load index and geoaccumulation index showed that sediments were unpolluted to extremely polluted by As. Therefore, sources of contamination to the Sabie River catchment should be closely monitored.

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