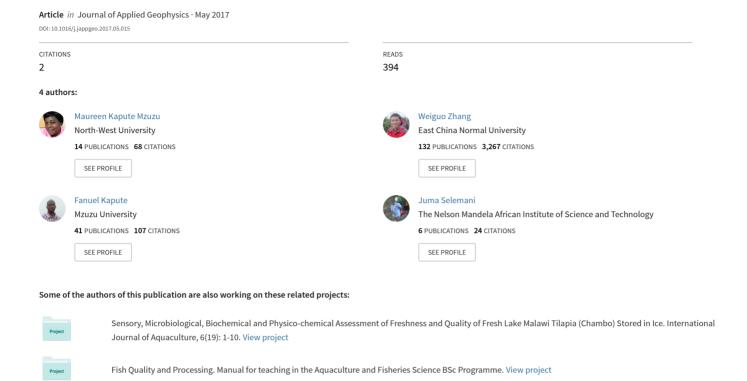
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Magnetic properties of sediments from the Pangani River Basin, Tanzania: Influence of lithology and particle size



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ABSTRACT

Use of magnetic properties in tracing sediment source in river systems is valuable for proper management of soil erosion and dam siltation. This study investigated magnetic properties variation from the upstream down to the coast of the Pangani River Basin in Tanzania, East Africa. The influence of lithology and sedimentary sorting on magnetic properties from source to sink pathway has been discussed. Results show that lithology can explain the differences in magnetic properties among the tributaries in the upstream and in the lower reach of the main stream. Ferrimagnetic minerals mainly consist of magnetite and titanomagnetite, with the latter more abundant in the upstream tributary Kikuletwa River. Upstream sediments have higher ferrimagnetic mineral concentrations but coarser grain size than the mainstream sediments, especially the dam sediments. A decline in ferrimagnetic mineral concentration with fining grain size is also observed along the mainstream. Such a trend can be explained by hydrodynamic sorting effect, which is well documented elsewhere. This study demonstrates that lithology and sorting have strong effect on magnetic properties, which should be properly addressed when using magnetic method in assessing fluvial sediment source.

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1. Introduction

Magnetic minerals (e.g., magnetite, hematite and goethite) are ubiquitous in soils and sediments (Thompson and Oldfield, 1986). Magnetic properties of sediments have been extensively applied worldwide in investigating sources of sediment entering rivers, lakes and oceans, in order to reconstruct environmental processes and establish countermeasures in heavily eroding areas of the catchment (Thompson and Oldfield, 1986; Zhang et al., 2008; Hatfield and Maher, 2009; Wang et al., 2011a; Collins et al., 2012; Nguyen et al., 2016; Pulley and Rowntree, 2016; Mzuza et al., 2017). Magnetic measurements have the advantage over most tracers in that measuring of magnetic properties is rapid, inexpensive, and non-destructive (Thompson and Oldfield, 1986; Walden et al., 1997). Lithology, sediment sorting and postdepositional processes have shown to affect magnetic mineralogy (Singer and Fine, 1989; Grimley et al., 2004; Walden et al., 1997; Owens et al., 1999; Walling, 2005; Fialová et al., 2006; Zhang et al., 2008; Wilkinson et al., 2013; Mzuza et al., 2017), and therefore, should be considered in sediment tracing using magnetic properties.

The Pangani River Basin is one of the largest and economically important water resources in Tanzania, East Africa, supplying water for

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agriculture, domestic purposes and production of electricity on the Nyumba ya Mungu Dam (NYM) (Ndomba et al., 2009). Pangani River drains its water into the Indian Ocean. There have been studies on Holocene environmental changes using sediment cores from the coast off Tanzania, in which mineralogical data are used to trace terrigenous sediment supply from the Pangani River (Liu et al., 2016). Geochemistry of bottom sediments of the Pangani River is used to study the lithological or anthropogenic source of elements (Hellar-Kihampa et al., 2012). However, no study in the Pangani River and Tanzania as a whole has been conducted on magnetic properties of sediments, which may provide a simple way for assessing sediment source.

In this study, magnetic properties of the Pangani River sediments along the transfer pathway from land to the ocean have been analyzed to understand the spatial variation of sediment compositions in terms of magnetic mineralogy. It is anticipated that the same set of magnetic parameters and measurements can be applied under various circumstances and provide certain general guidelines in interpreting magnetic data in terms of lithological and particle size influence.

2. Study area

The Pangani River Basin (PRB) is located between coordinates 36°20′ E, 02°55′ S and 39°02′ E, 05°40′ S in part of Tanzania (PBWO/IUCN, 2007; Ndomba et al., 2009) (Fig. 1). The river basin covers an area of about 43, 650 km² (Turpie et al., 2005), and is one of the largest

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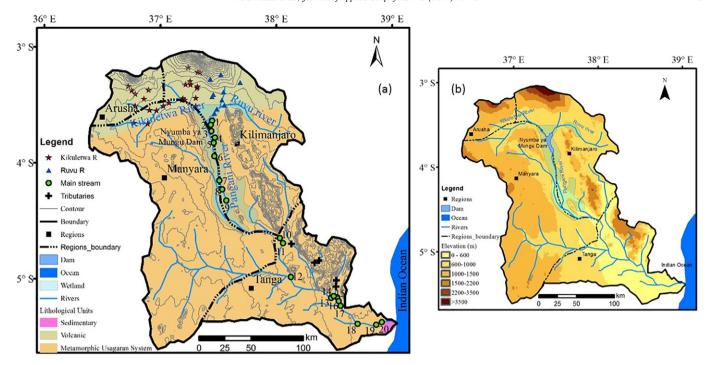


Fig. 1. Geological (a) and topographic (b) map of the catchment area of the Pangani River basin, Tanzania. Symbols represent sampling sites, Red stars for Kikuletwa River sites, blue triangles for Ruvu sites, green circles for the main stream and black cross for some tributaries in the mainstream (Source: IUCN, 2009).

River Basins along the Tanzanian Indian Ocean drainage system (Pamba et al., 2016). Five percent of the Pangani River Basin is located in Kenya but this study covered the Tanzanian part only. The basin covers part of Kilimanjaro, Arusha, Manyara and Tanga regions, supporting a population of about 6 million people (National Bureau of Statistics, 2013). The Pangani River system rises into the southern and eastern sides of Africa's highest peak, Mt. Kilimanjaro (5985 m) and Mt. Meru (4566 m), which together create the Kikuletwa and the Ruvu tributaries. The two tributaries join at Nyumba ya Mungu Dam which is 150 km², 40 m deep and has a storage capacity of about 875 million m³ (Shaghude, 2006; Hellar-Kihampa et al., 2012) (Fig. 1). After flowing out from the dam, the main stream of Pangani River passes through the arid lowlands also known as the *Maasai steppe*, and draining the Pare and Usambara Mountains, before reaching the coast (IUCN, 2003; Ndomba et al., 2009; Hellar-Kihampa et al., 2013a).

The Pangani River Basin experiences a tropical climate where rainfall increases with altitude, and highlands and lowlands receive more rainfall (between 1000 and 2000 mm) and (between 500 and 600 mm) respectively per year (Rohr and Killingtveit, 2003; PBWO/IUCN, 2008; Hellar-Kihampa et al., 2012). Due to north-southward movement of the inter-tropical convergence zone (ITCZ), the basin has a bimodal type of rainfall, where the long rainy season occurs between March to May and short rainy season from November to December (Kijazi and Reason, 2009; Mahongo and Shaghude, 2014). Minimum water discharge in Pangani River usually occurs between December and February (20 m³/s) while maximum discharge (97 m³/s) is usually in May (Pamba et al., 2016). According to Ndomba et al. (2009), annual sediment yield rate for Kikuletwa River catchment is 0.419 Mt./year, while Ruvu is 11,000 t/year. It is reported that NYM Dam releases sediment load of about 7939 t/year (Mtalo and Ndomba, 2002; Ndomba et al., 2009; Ndomba, 2010). The highlands experience low temperature, while the larger part of the basin experiences high temperature throughout the year. Generally, temperature ranges from 14 °C to 35 °C. Lowest temperatures (14-18 °C) are experienced between July and August while January to February are warmer months (32–35 °C) (PBWO/IUCN, 2008). The area above the dam (upstream) is volcanic consisting of olivine and alkaline basalts, phonolites, trachytes, nephelinites and pyroclastics (Schluter, 2008; PBWO/IUCN, 2008). The central region of the Pangani River Basin is characterized by Proterozoic crystalline rocks which consist of granulitic gneisses (Sommer et al., 2003; PBWO/IUCN, 2008). The coastal region is dominated by Mesozoic and younger sediments (Liu et al., 2016). The soils in the area above NYM Dam are mainly Chernozem, Leptosols and Luvisols for the Kikuletwa River catchment, and Cambisols, Histosols and Andosols for the Ruvu River catchment (PBWO/IUCN, 2008).

3. Materials and methods

Sediment samples were collected in two seasons, wet and dry in 2014 and 2015, respectively. Sampling locations were distributed throughout the catchment (Fig. 1). Samples were collected from above the NYM Dam which includes Kikuletwa (27) and Ruvu (10) Rivers, the NYM Dam, and main stream (Pangani River) to the coast (20), including some tributaries (Mkomazi, Soni and Luengera) below NYM Dam (5).

All samples were dried at 40 °C and then disaggregated before analysis. Particle-size distribution was measured using a laser-diffraction analyser (Coulter LS-100Q) following pre-treatment with 0.2 M HCl and 5% H₂O₂ to remove biogenic carbonate and organic matter, respectively. Sodium hexametaphosphate [Na (PO₃)₆] was added to the sample and ultra-sonicated to ensure complete disaggregation before analysis (Ru, 2000).Low-frequency (0.47 kHz) and high-frequency (4.7 kHz) magnetic susceptibility (χ lf and χ hf, respectively) were measured using a Bartington magnetic susceptibility meter and MS2B dual-frequency sensor. Frequency-dependent susceptibility ($\chi_{fd\%}$) was calculated as $\chi_{fd\%} = (\chi lf - \chi hf) / \chi lf \times 100$. Anhysteretic remanent magnetization (ARM) was imparted in a 0.04 mT direct current field superimposed on a peak AF demagnetization field of 100 mT, and is expressed as susceptibility of ARM (χ_{ARM}) by normalizing ARM with DC field. Isothermal Remnant Magnetization (IRM) measurements were made using a forward field of 1 T followed by backfields of -100 mT and -300 mT. The corresponding IRM is referred to as

Table 1 Sediment particle size composition and magnetic characteristics (mean \pm SD, Minimum and Maximum value.

Parameter	Above dam ($n = 37$)						Main stream ($n = 20$)			Tributaries below dam $(n = 5)$		
	Kikuletwa River ($n = 27$)			Ruvu River ($n = 10$)								
	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max
Clay (%)	18 ± 9	7	35	21 ± 11	3	37	20 ± 11	5	44	14 ± 6	7	21
Silt (%)	32 ± 10	18	58	31 ± 17	7	60	46 ± 20	11	75	24 ± 10	15	38
Sand (%)	50 ± 17	16	75	48 ± 28	11	90	34 ± 28	2	84	62 ± 16	44	78
$\chi (10^{-8} \text{m}^3 \text{kg}^{-1})$	1379 ± 536	139	2293	820 ± 709	189	2150	144 ± 108	28	425	154 ± 62	70	241
χ _{fd%} (%)	0.8 ± 0.6	0.0	2.6	1.4 ± 1.2	0.2	3.6	2.6 ± 2.0	0.3	8.0	3.3 ± 3.0	0.4	8.5
$\chi_{ARM} (10^{-8} \text{m}^3 \text{kg}^{-1})$	4158 ± 1436	1849	7875	4495 ± 3371	503	8701	747 ± 549	135	1883	534 ± 211	299	836
SIRM $(10^{-6} \text{Am}^2 \text{kg}^{-1})$	$152,250 \pm 55,603$	74,010	314,489	$138,701 \pm 127,046$	15,109	342,039	$14,101 \pm 12,421$	2670	48,759	$10,336 \pm 8660$	4718	25,702
HIRM $(10^{-6} \text{Am}^2 \text{kg}^{-1})$	1708 ± 1263	342	5720	1329 ± 1046	375	2938	333 ± 181	83	781	463 ± 302	259	991
S ₋₁₀₀ (%)	92.3 ± 1.7	89.6	95.1	89.5 ± 3.4	84.7	93.3	87.7 ± 4.4	75.5	92.2	81.9 ± 3.3	78.9	86.8
S ₋₃₀₀ (%)	98.7 ± 1.2	95.2	99.8	98.2 ± 1.2	96.2	99.8	96.8 ± 2.1	92.0	98.9	95.0 ± 1.5	92.9	96.1
$\chi_{ARM}/SIRM~(10^{-5}~mA^{-1})$	28 ± 6	16	42	43 ± 24	21	98	57 ± 21	23	105	70 ± 35	17	110

IRM $_{\chi mT}$, where χmT indicates the field value. IRM obtained at 1 T was referred to as saturated IRM (SIRM). Hard IRM (HIRM) was defined as HIRM = 0.5 × (SIRM + IRM $_{-300mT}$). S $_{-100}$ and S $_{-300}$ were calculated as S $_{-100}$ = 0.5 × (SIRM-IRM $_{-100mT}$) / SIRM × 100 and S $_{-300}$ = 0.5 × (SIRM – IRM $_{-300mT}$) / SIRM × 100, respectively (Zhang et al., 2008).

Measurements of the temperature-dependent magnetic susceptibility were made using an AGICO MFK1-FA Kappabridge equipped with a CS-3 high-temperature furnace. Each sample was heated from room temperature to 700 $^{\circ}\text{C}$ and then cooled to room temperature in an argon atmosphere.

4. Results

4.1. Magnetic properties

4.1.1. Upstream and main stream comparison

The upstream samples include Kikuletwa and Ruvu Rivers while the mainstream samples start from NYM Dam down to the Indian Ocean coast (Table 1). Generally, magnetic susceptibility χ and SIRM reflects the concentration of magnetic minerals. SIRM is not influenced by (super) paramagnetic and diamagnetic minerals unlike χ (Thompson

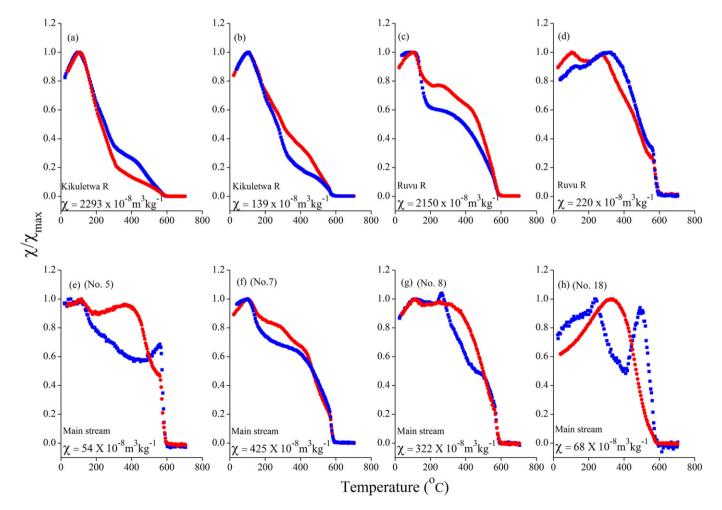


Fig. 2. Magnetic susceptibility versus temperature curves for upstream (a-d) and mainstream (e-h) sediment. Red and blue lines represent heating and cooling curves, respectively.

and Oldfield, 1986). χ_{ARM} is sensitive to stable single domain (SD) of between 0.04 and 0.06 mm ferrimagnetic grains (Maher, 1988). HIRM is normally used to estimate abundance of imperfect anti-ferromagnetic minerals such as goethite and hematite (Bloemendal and Liu, 2005; Nguyen et al., 2016). The upstream sediments had higher χ , χ_{ARM} , HIRM and SIRM values, which are 8, 6, 10 and 5 times higher respectively, than the mainstream sediment, suggesting higher ferrimagnetic mineral concentrations (Table 1). Tributary values were almost similar to the main stream values (Table 1).

 $\chi_{fd\%}$ estimates relative contribution of fine, viscous superparamagnetic (SP \sim <0.03 μm) for magnetite grains to the total magnetic assemblage (Thompson and Oldfield, 1986). $\chi_{fd\%}$ was relatively higher in the mainstream samples than in the upstream (Table 1), suggesting higher proportions of SP magnetite grains in the former. The ratio of $\chi_{ARM}/SIRM$ was also indicative of grain-size, and not affected by SP particles. Therefore, lower values correspond to coarser magnetic domain (MD) grains. Lower $\chi_{ARM}/SIRM$ ratios for upstream samples may suggest coarser grain size in sediment of upstream samples (Oldfield, 1994) (Table 1).

According to Bloemendal and Liu (2005), S_{-300} measures relative importance of low-coercivity (e.g. magnetite and maghemite) and high-coercivity components in the total magnetic mineral assemblage, while S_{-100} reflects the ratio of low-coercivity minerals to medium-coercivity and high-coercivity minerals (Yamazaki, 2009; Yamazaki

and Ikehara, 2012). All samples had S_{-300} values >95%, implying a dominance of magnetic properties by ferrimagnetic minerals (Thompson and Oldfield, 1986) (Table 1). Mean values for parameters indicating magnetic mineralogy (S_{-100} , S_{-300}) were slightly higher in the upstream samples (Table 1), suggesting higher proportions of ferrimagnetic minerals in the upstream.

Thermomagnetic analysis revealed Curie temperature around 600 °C in all the samples, confirming that magnetite is the dominant ferrimagnetic mineral (Fig. 2). In addition, the samples generally showed a Curie temperature around 200 °C and 350 °C, indicating the presence of titanium (Ti) containing magnetite (Thompson and Oldfield, 1986) (Fig. 2). The ~350 °C Curie temperature signal was more pronounced in upstream sediments of Kikuletwa River (Fig. 2a, b) than those from the Ruvu River (Fig. 2c, d), suggesting that titanium content is higher in sediments of the former. The dam sample (Fig. 2e) and other main stream samples (Fig. 2f-h) showed thermomagnetic pattern much similar to those of the Ruvu River samples (Fig. 2c–d).

There was a positive correlation between χ and SIRM for all the samples, although the relationships were not as good for the upstream samples compared to those from the mainstream (Fig. 3a). This suggests that χ is mainly contributed by ferrimagnetic minerals. The positive correlation trend was also observed between χ and χ_{ARM} and χ and HIRM (Fig. 3b, c). Negative correlation was observed between χ and grain-size indicators (χ_{ARM} /SIRM and χ_{ARM} / χ) for the upstream samples

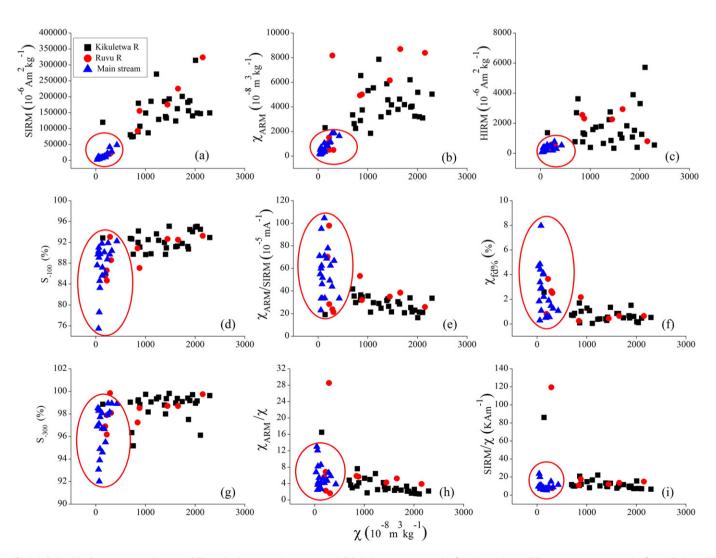


Fig. 3. Relationships between magnetic susceptibility and other magnetic parameters. Solid circles represent samples from Ruvu River, solid squares represent samples from Kikuletwa River and triangular in red circles represent main stream samples.

(Fig. 3e, h), implying that sediments containing higher ferrimagnetic mineral concentrations have a coarser magnetic grain size. Half of Ruvu River samples (5 samples) were also within the main stream range in all the parameters (Fig. 3).

4.1.2. Spatial variations within the upstream and along the main stream

Magnetic mineral concentration parameters (χ , SIRM and χ_{ARM}) and S-ratios (S_{-100} , S_{-300}) were higher in the Kikuletwa samples (Fig. 4). On the contrary, grain-size indicators (χ_{ARM} /SIRM and $\chi_{fd\%}$) were relatively higher in the Ruvu River samples (Fig. 4e and f), while χ_{ARM}/χ was generally similar in both river sites (Fig. 4g). Mainstream, magnetic mineral concentration parameters (χ , SIRM, HIRM and χ_{ARM}) were lowest in the dam samples (Fig. 5a–d). There was an increase in magnetic mineral concentration parameters downstream of the dam especially for samples within the marsh area (Fig. 5, no. 6 - no. 9), which decreased towards the coast (Fig. 5a–d). χ_{ARM} /SIRM and SIRM/ χ increased and decreased respectively towards the coast (Fig. 5e, h).

4.2. Particle size distributions

Particle size composition analysis on spatial variation showed that upstream samples have coarser sediment than the mainstream sediment (Table 1, Fig. 6). On average, upstream sediment had 19% clay ($<4 \mu m$), 32% silt (4–63 μm) and 49% sand ($>63 \mu m$), while mainstream

sediment had 20% clay, 46% silt and 34% sand (Table 1, Fig. 6). There was a decreasing trend in particle size distribution in the mainstream (Fig. 6d–f) from the dam to the coast. The first 4 dam samples had very low sand percentage of <2% (Fig. 6f).

4.3. Correlation between sediment particle size and magnetic properties

Correlation analysis showed different relationships between particle size and magnetic properties (Table 2). χ and SIRM were positively correlated with the fraction 250–500 μ m (p < 0.05) (Table 2). $\chi_{ARM}/SIRM$ was positively correlated with the fraction <4 μ m, 4–16 μ m and 16–63 μ m (Table 2) while χ_{ARM}/χ was positively correlated with the fraction <4 μ m and 4–16 μ m (Table 2). S-ratios were positively correlated with fraction <4 μ m and 4–16 μ m (Table 2).

5. Discussion

5.1. Lithological influence on magnetic properties of sediments

Post-depositional reductive diagenesis has influence on magnetic properties, but our surface samples were collected along the river banks, which is in frequently contact with atmosphere. It is expected that the samples were subjected to oxidizing environment, and therefore reductive diagenesis is not important. Some of the samples are from the dam, and the water in the dam is oxic (Hellar-Kihampa et al.,

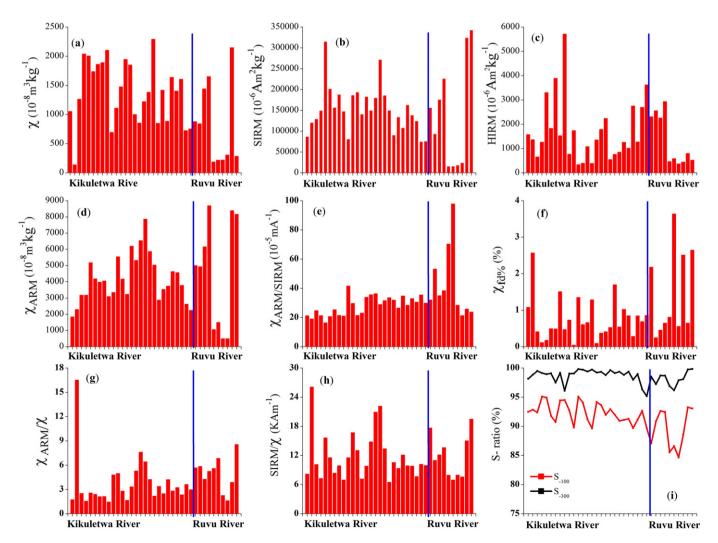


Fig. 4. Spatial variation of magnetic parameters within the upstream. The blue line separates the two river sampling sites (Kikuletwa and Ruvu River).

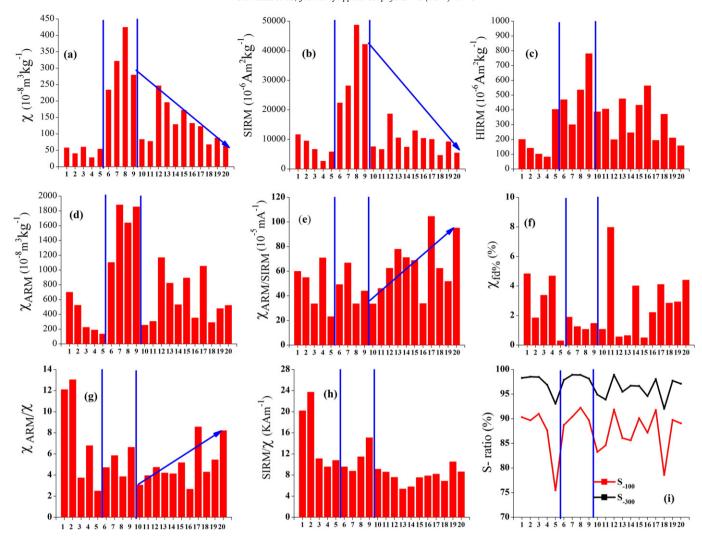


Fig. 5. Spatial variation of magnetic parameters from the dam down to the coast. The zone between the two vertical lines demarcate the wetland samples with higher magnetic characteristics.

2013a), and therefore the surface sediments in the dam are not heavily altered by reductive diagenesis. Therefore, changes in lithology between the upstream and main stream could be the main reason for differences in magnetic properties (Fig. 1). Volcanic mountains of Kilimanjaro and Meru are located in the northern part of the Pangani River Basin above the NYM Dam (Hellar-Kihampa et al., 2013b). The upstream region is covered by Tertiary-Quaternary volcanic rocks including basalts, nephelinites and photonites (PBWO/IUCN, 2007), which are magnetite rich rocks (Thompson and Oldfield, 1986). The region of the mainstream is mainly composed of Precambrian metamorphic rocks and sedimentary rock (PBWO/IUCN, 2007) (Fig. 1). In comparison to the volcanic rocks in the upstream, these rocks from the main stream are magnetically weaker (Thompson and Oldfield, 1986), and hence, sediments derived from this region should be magnetically weaker. Therefore, upstream sediments show much stronger χ and SIRM values in comparison to tributaries of the mainstream sediments (Table 1).

Magnetic properties of sediments in the upstream also show differences between the two rivers which could be due to local variations in rock types, as the sediments from the Kikuletwa River show higher proportions of titanomagnetite (Fig. 2). Previous geochemical study also showed that sediments from the Kikuletwa River contain higher Ti content than other sites in the Pangani River basin (Hellar-Kihampa et al., 2012). Considering the multiple volcanic activities of different ages in

the study area (Nonnotte et al., 2008), it is expected that sediments within the upstream show significant variations of magnetic properties (Wang et al., 2011b). It is interesting to note that the mainstream sediments show thermomagnetic curve more close to that of the Ruvu River sediments (Fig. 2). It may be suggested that Ruvu River has an important sediment contribution to the mainstream. Such assertion nevertheless, needs further study, including sediment budget and geochemical analyses.

5.2. Particle size control

Findings in this study agree with earlier studies that magnetic properties of bulk sediment show dependence on particle-size compositions (Hatfield and Maher, 2009; Gallaway et al., 2012; Dong et al., 2014). It seems that ferrimagnetic minerals are largely concentrated in sand fraction as shown by the positive correlation coefficients between χ , SIRM and 250–500 μ m fraction (Table 2). As a result, the upstream sediments are coarser and show higher mineral magnetic content compared to the mainstream sediments of finer particle size (Table 1 and Fig. 6).

The lower values of χ_{ARM}/χ (<10) and $\chi_{ARM}/SIRM$ (<20 × 10⁻⁵ mA⁻¹) for the upstream sediments indicate that magnetic minerals are dominated by coarse pseudo-single domain and multi-domain grains (Oldfield, 1994; Li et al., 2015), while the mainstream sediments have

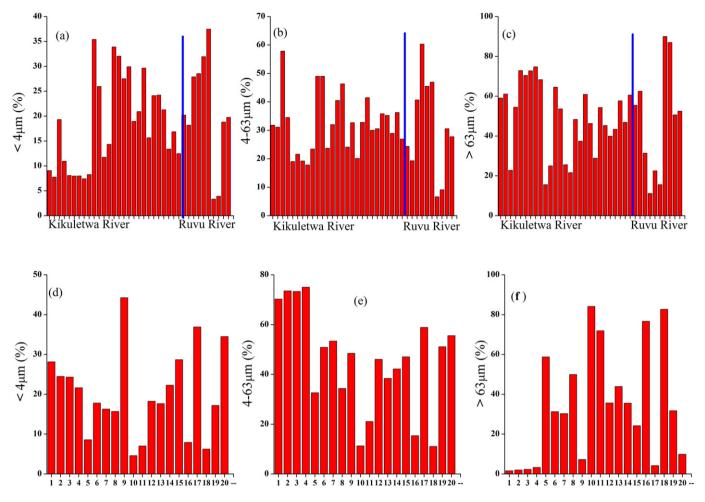


Fig. 6. Spatial variation of particle-size composition of the upstream (a-c) and mainstream (d-f) sediments. Line demarcates samples from Kikuletwa and Ruvu Rivers in the upstream.

higher χ_{ARM}/χ and $\chi_{ARM}/SIRM$ values and therefore, containing higher proportions of fine-grained ferrimagnetic minerals (Table 1 and Fig. 6). During sediment transport from upstream down to the coast, coarse ferrimagnetic minerals will preferentially be removed from the transferring pathway and therefore, sediments in the downstream will be reduced in magnetic mineral concentration and become finer (Hatfield and Maher, 2009; Wang et al., 2010; Gallaway et al., 2012; Dong et al., 2014; Nguyen et al., 2016). Lower magnetic mineral concentration but finer grain size in mainstream sediments in comparison to upstream sediments is therefore likely due to the sorting effect (Fig. 5a-d). Lowest values of χ and SIRM in the dam samples are in accordance with the lowest sand fractions therein (Fig. 5a-d). The increase in magnetic concentration in the wetland area downstream of the dam (Fig. 1 sample No. 6–9) can be explained by the increase of coarser particle size fractions (Fig. 5a-d). With the continuous

deposition of coarse ferrimagnetic minerals downstream, χ and SIRM show decreasing trend, while $\chi_{ARM}/SIRM$ shows the opposite - a good indication of sorting effect on magnetic properties.

6. Summary and conclusions

Magnetic properties of sediments in the upstream of the Pangani River are dominated by magnetite and titanomagnetite, and the latter is more abundant in the Kikuletwa River catchment, reflecting the spatial variation of volcanic rock types. Upstream sediments show higher ferrimagnetic mineral concentrations but coarser grain size, while the mainstream sediments have lower ferrimagnetic mineral content but finer grain size, which can be explained by sorting effect during the source to sink transport of river sediments. This study demonstrates that magnetic properties of sediments are sensitive to lithology and

Table 2 Correlation coefficients between magnetic properties and particle size (n = 62).

Particle size	χ	SIRM	HIRM	χfd%	χarm	χarm/SIRM	χarm/χ	S ₋₁₀₀	S ₋₃₀₀
< 4 μm	-0.08	0.02	-0.18	-0.02	0.21	0.42**	0.28*	0.23	0.32*
4–16 μm	-0.12	-0.10	-0.20	0.09	-0.01	0.26*	0.27*	0.25*	0.31*
16-63 μm	- 0.31 *	- 0.33**	-0.34**	0.13	-0.27^*	0.43**	0.16	0.10	0.14
63-250 μm	0.02	-0.04	-0.10	0.15	-0.21	-0.22	-0.30*	-0.23	-0.28*
250-500 μm	0.30*	0.28*	0.25	-0.18	0.09	-0.44**	-0.23	-0.13	-0.16
500-2000 μm	0.13	0.12	0.44**	-0.19	0.18	-0.24	-0.04	-0.10	-0.16
Mean size	0.17	0.15	0.43**	-0.17	0.16	-0.32*	-0.11	-0.14	-0.20

^{*} Significant at p < 0.05.

^{**} Significant at p < 0.01.

hydrodynamic sorting, which is useful in characterizing the spatial variation of sediment composition within a river basin.

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