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Geochemical characteristics of stream sediments, sediment fractions, soils, and basement rocks from the Mahaweli River and its catchment, Sri Lanka

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ABSTRACT

Geochemical variations in stream sediments ($n = 54$) from the Mahaweli River of Sri Lanka have been evaluated from the viewpoints of lithological control, sorting, heavy mineral concentration, influence of climatic zonation (wet, intermediate, and dry zones), weathering, and downstream transport. Compositions of soils ($n = 22$) and basement rocks ($n = 38$) of the catchment and those of <180 μm and 180–2000 μm fractions of the stream sediments were also examined. The sediments, fractions, soils and basement rocks were analyzed by X-ray fluorescence to determine their As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc, Fe_2O_3, TiO_2, MnO, CaO, P_2O_5 and total sulfur contents. Abundances of high field strength and ferromagnesian elements in the sediments indicate concentration of durable heavy minerals including zircon, tourmaline, rutile, monazite, garnet, pyriboles, and titanite, especially in <180 μm fractions. The sediments show strong correlation between Ti and Fe, further suggesting presence of heavy mineral phases containing both elements, such as ilmenite and magnetite. The basement rocks range from mafic through to felsic compositions, as do the soils. The river sediments lack ultrabasic components, and overall have intermediate to felsic compositions. Elemental spikes in the confluences of tributary rivers and high values in the <180 μm fractions indicate sporadic inputs of mafic detritus and/or heavy minerals to the main channel. $Al_2O_3/(K_2O + Na_2O)$ and K_2O/Na_2O ratios of the sediments and LOI values of the soils correlate well with the climatic zones, suggesting intense weathering in the wet zone, lesser weathering in the intermediate zone, and least weathering in the dry zone. Low Sr and CaO contents and Cr/V ratios in stream sediments in the wet zone also suggest climatic influence. Fe-normalized enrichment factors (EFs) for As, Pb, Zn, Cu, Ni and Cr in stream sediments in the main channel are nearly all <1.5, indicating there is no significant environmental contamination. The chemistry of the sediments, rocks and the soils in the Mahaweli River are thus mainly controlled by source lithotype, weathering, sorting, and heavy mineral accumulation.

1. Introduction

Geochemical investigations based on the chemical analysis of active stream sediments are an effective tool with multiple applications (Grunsky et al., 2009). Such geochemical surveys were initially used as an exploration tool, but their application has now evolved into a more extensive technique, particularly for environmental purposes (Ranasinghe et al., 2008; Grunsky et al., 2009). The geochemical compositions of stream sediments reflect the average composition of an entire drainage basin (Reimann and Melezhik, 2001; Halamic et al., 2001). Although the combined effects of chemical, biological and physical weathering processes transform bedrocks into soils and ultimately into sediments, original geochemical signatures of the source may be retained (Formoso, 2006). Trace elements such as Sc, Th, Zr, Cr, Ni and Co are generally immobile during surficial processes (Taylor and McLennan, 1985), and their abundances in sediments are thus useful indicators of source composition. However, consideration of whole-rock and soil chemistry gives a more comprehensive overview of the processes that have operated during the production of sediments. Consequently, considering data for soils and source rocks in combination with data for stream sediments is advantageous.

Few geochemical studies of stream sediments have been carried out in Sri Lanka, and most of those conducted to date have focused on mineral exploration (Dept. of Mineralogy, 1959; Dissanayake and Rupasinghe, 1992; Gamage et al., 1992; Fernando, 1995; Ruapasinghe, 2000; Dissanayake and Chandrajith, 2003; Ranasinghe et al., 2009). Dissanayake and Chandrajith (2003) examined stream sediments in the Walawe basin, which covers part of the Highland and Vijayan complexes (Fig. 1). The Walawe basin is not subject to variable climatic influence, as it lies entirely

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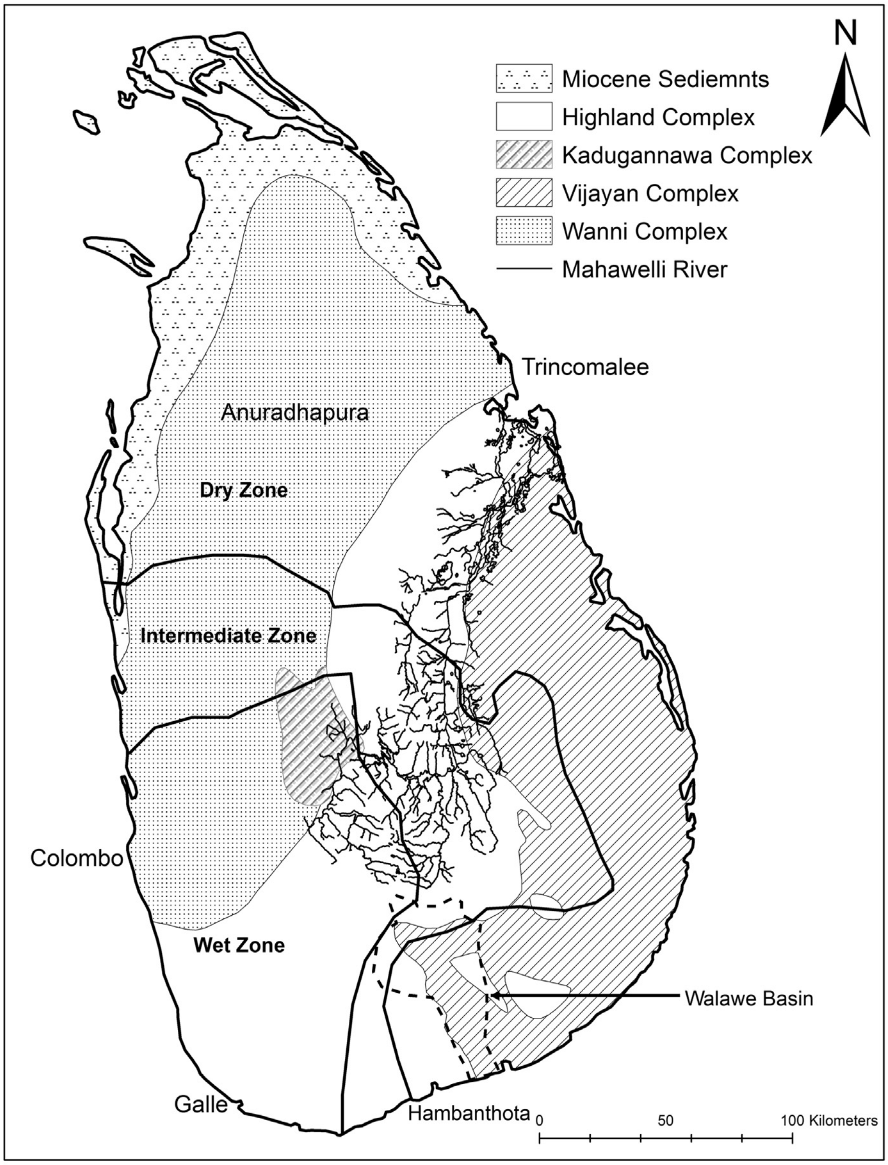

Fig. 1. Simplified geological and climatic map of Sri Lanka (after Cooray, 1994) showing location of the Mahaweli River and the Walawe Basin.

within the wet zone of Sri Lanka. In contrast, the Mahaweli River, the focus of our present study, transects the three climatic zones (wet, intermediate and dry) of Sri Lanka.

The Mahaweli River is about 335 km in length, and enters the Indian Ocean in Trincomalee Bay. The Mahaweli River receives sediment from many tributaries that flow across metamorphic basement, resulting in mixing of detrital components. A feature of this river is that it has relatively few tributaries in its lower reaches, whereas many tributaries are found in its upper reaches (Fig. 2).

This study examines the effects of provenance, climate, heavy mineral accumulation, grain size, and hydraulic sorting in Mahaweli River sediments, using selected major and trace elements. Data for bulk composition (<2000 μm) and two size fractions (<180 μm and 180–2000 μm) are employed for the stream sediments, along with whole rock data for source rocks and soils. In addition, the EF (enrichment factor) for Pb, Zn, Cu, Ni, Cr and As contents have been calculated using Fe as the reference element, to identity any potential environmental effects.

2. General geology, climate and the study area

The drainage basin of the Mahaweli River is the largest in Sri Lanka at 10,448 km^2, covering almost one-sixth of the country. The Mahaweli basin has a trellised drainage network within which the main channel and its extensive floodplain receive inputs from a series of tributaries of varying size (Ranasinghe et al., 2008).

Sri Lanka mainly consists of high-grade Precambrian metamorphic rocks divided into three major lithotectonic units (Fig. 1), the Highland, Vijayan, and Wanni complexes. The largest of these is the Highland Complex, which is made up of supracrustal rocks and a variety of igneous intrusions. It forms the backbone of the Precambrian rocks of N–S trending granulite facies metamorphites, and consists predominantly in varieties of granulites, including charnockites, quartz-feldspar-garnet-sillimanite-graphite schists, quartzites, marbles, metabasites, and calc-gneisses (Pohl and Emmermann, 1991; Cooray, 1994). The headwaters of the Mahaweli River lie mostly within the Highland Complex (Fig. 1). However, the current study deals with sediments derived only

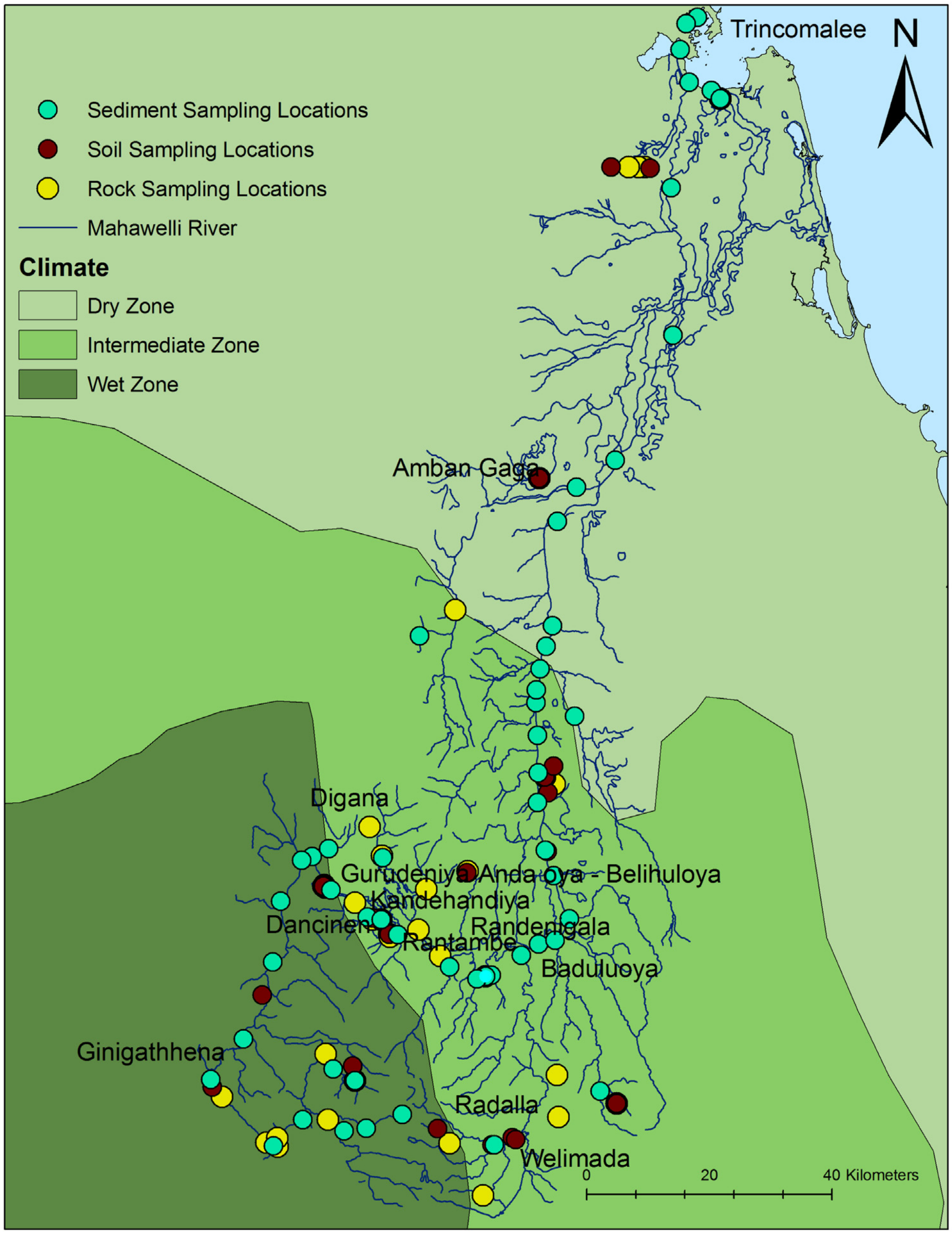

Fig. 2. Location of sediment, rock, and soil sample sites along the Mahaweli River and distribution of the wet, intermediate and dry climatic zones in the study area.

from the Highland Complex, and geochemical variations along the Mahaweli River.

Sri Lanka is divided into three climatic zones (wet, intermediate and dry) based on the amount of precipitation received (Fig. 1). The Mahaweli flows through all three zones. The wet zone receives rain from both the SW and NE monsoons, as well as the inter-monsoon period (March to September). The dry zone receives rain only during the NE Monsoon between the months of October and March. The intermediate zone experiences rainfall between the dry and the wet zone extremes, and receives rain from all monsoons.

3. Methods

Stream sediment samples were collected along the Mahaweli River, taking into account sediment deposition, flow rate, river gradient, erosion and human impact on natural sedimentation. Samples were taken at varying distances based on accessibility (Fig. 2), considering locations of tributaries, geological factors, and the distribution of the three climatic zones. Four to five kg bulk samples were taken at each location.

The stream sediment samples were oven dried at 120 °C prior to geochemical analysis at Shimane University. Samples were first hand-sieved to remove the small amount of material >2000 μm. The <2000 μm fraction was then split to provide a sample for bulk sediment analysis (<2000 μm). A second split was then sieved to separate <180 μm and 180–2000 μm fractions for fractional analysis. The <180 μm and 180–2000 μm fractions are often used as the most appropriate and convenient fractions for the examination of stream sediment geochemistry (Amorosi et al., 2002; Ferreira et al., 2001; Koval et al., 1995; Licht and Tarvainen, 1996; Ortiz and Roser, 2006a,b). Consequently, the <180 μm and 180–2000 μm fractions were also used for this study.

Whole-rock samples were cut into small chips and washed with distilled water, ultrasonically cleaned for 10 min to remove clay particles, again rinsed with distilled water, and then oven dried at 110 °C for 48 h. Approximately 50 g of each fraction, bulk sediment and soil samples were oven-dried at 160 °C for 48 hrs before crushing. All samples were crushed using a ROCKLABS tungsten carbide ring mill, with mill times of <60 s.

The crushed sediment, soil and rock samples were compressed into briquettes, using a force of 200 kN for 60 s. The concentrations of 19 major and trace elements were then determined by X-ray florescence spectrometry using a Rigaku RIX-2000 spectrometer equipped with a Rh-anode tube. Analytical methods, instrumental conditions, and calibration followed those described by Ogasawara (1987). Major element analyses of <180 μm and 180–2000 μm fractions of the river sediments

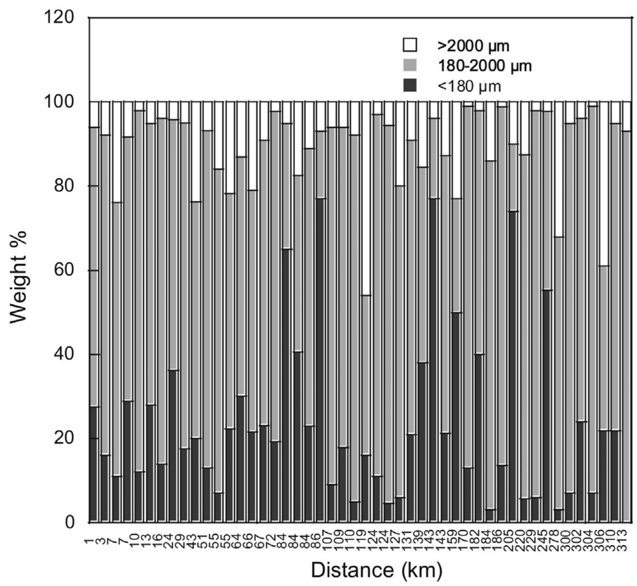

Fig. 3. Variation in stream sediment size fractions along the Mahaweli River.

were also made using glass fusion beads (Kimura and Yamada, 1996). These data were used for calculation of bulk Al_2O_3, K_2O and Na_2O contents used in preliminary evaluation of weathering (Section 5.4).

Heavy mineral separates of selected samples were prepared using conventional heavy liquid and magnetic separation (Frantz Isodynamic L-1), followed by counting using a binocular microscope (20–40×).

4. Results

4.1. Textural and mineralogical characteristics

The main channel sediments were coarse to medium sand with lesser clay fractions, except for eleven samples in the middle reaches that contained more than 20% clay (Fig. 3). The gravel content of most samples was low (<20%), except for one sample in the middle reaches and two in the lower reaches.

In general, sediment grain size in rivers decreases downstream (Parker, 1991; Surian, 2002) but this is not the case for the Mahaweli. There is little improvement in sorting along most of the main channel, and median grain size is also highly variable. This is probably due to the number of tributaries entering the main channel, even in the middle reaches. The sediments only become well sorted in the lowermost reaches of the river, where tributaries are fewer.

The main minerals present in the stream sediments include quartz, plagioclase, K-feldspar, muscovite, hornblende and biotite. Based on visual estimates, the stream sediments in the upper reaches are richer in feldspar, whereas quartz content increases in the lower reaches. Common minor or accessory minerals include garnet, sillimanite, monazite, zircon and apatite. The river and its tributaries run along or across basement rocks including charnockitic gneiss, marble, hornblende-biotite gneiss, quartzite, garnet-sillimanite gneiss and metabasite. Microscope examination of the heavy mineral separates showed that zircon, magnetite, ilmenite, garnet, tourmaline, hornblende and allanite were the most abundant heavy minerals, whereas biotite and apatite were less abundant.

The soil samples collected were mostly pale loamy to clayey residual soils. A few samples that were rich in organic matter were darker in hue, and others were pink to red. The basement rocks sampled represent a variety of lithotypes. These included

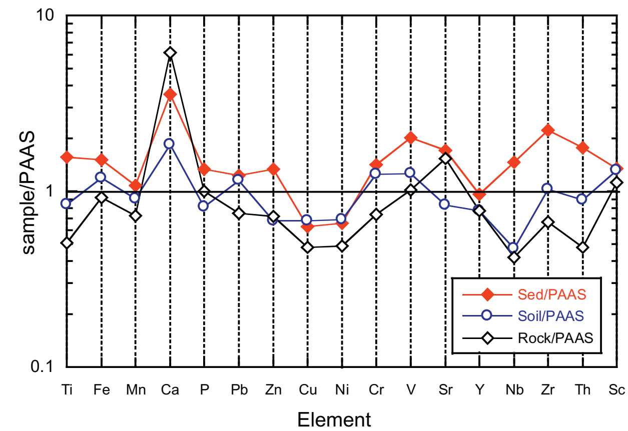

Fig. 4. PAAS-normalized averages for the sediments, soils and basement rocks of the Mahaweli River. PAAS values from Taylor and McLennan (1985). Major elements are normalized as oxides.

medium- to coarse-grained charnockites, granitic gneisses, marbles, meta-pelites, quartzites, and metabasites.

4.2. Data description

Individual analyses and averages of the bulk stream sediments, rocks and soils are given in Tables 1a, 1b and 1c, respectively, as supplementary data. Data for the <180 µm and 180–2000 µm sediment fractions will be available in a future publication, but can be provided on request. Averages and minimum and maximum values are summarized in Table 2. Average values of Fe_2O_3, TiO_2, MnO and P_2O_5 are highest in the river sediments and lowest in the source rocks, whereas average CaO is highest in the source rocks and lowest in the soils (Table 2). Many of the trace elements (Zn, Cr, V, Sr, Y, Nb, Zr, Th, TS) have highest average values in the river sediments, which also tend to record the highest maximum values.

In order to highlight the differences between the sample types and to compare with typical upper crustal values, average values in the sediments, rocks and soils were normalized against average post-Archean Australian shale (PAAS; Table 2, Taylor and McLennan, 1985). The river sediments are highly enriched in CaO, V and Zr, and moderately enriched in TiO_2, Cu, Cr, Sr, Nb, Th and Sc relative to PAAS (Fig. 4). Several elements in the sediments (Pb, MnO and Y) exhibit PAAS-like values, and Cu and Ni are depleted. The sediments are slightly enriched in P_2O_5 relative to the soils and rocks, possibly due to concentration of accessory phases such as apatite and monazite (Nagarajan et al., 2007).

Most elements in the soils are depleted (e.g. Zn, Cu, Ni, Y and Nb), whereas CaO is moderately to highly enriched relative to PAAS. The rocks are also mostly depleted relative to PAAS, especially for TiO_2, Cu, Ni, Nb, Zr and Th. In contrast, CaO and Sr are enriched.

Titanium is relatively immobile compared to other elements during sedimentary processes, and hence is a good indicator of source rock composition (McLennan et al., 1993); it is also mainly concentrated in phyllosilicates (Condie, 1992). Average TiO_2 content in the basement rocks is depleted relative to PAAS, suggesting they are more evolved (felsic) than typical upper continental crust (Nagarajan et al., 2007). Enrichment of CaO in the rock average (5.82 wt%) is due to inclusion of five marbles in the dataset. If these samples are removed, average CaO falls to 3.10 wt%, only double that of PAAS (1.29 wt%). High Sr values (>1000 ppm) in three gneisses also contribute to the enrichment in Sr in the basement rocks relative to PAAS. Overall, the rocks and the soils are mainly depleted relative to PAAS, whereas the sediments are mainly enriched. This indicates that geochemical changes have occurred in the steps from source to sediment.

Table 1a
Analyses of Mahaweli River bulk stream sediments. Major elements as oxide wt%, trace elements ppm. Distance: distance downstream (km) from the uppermost sample point. The highlighted rows are the tributary values.

S-no.	Distance (km)	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5
Wet zone																				
2	1	10	26	165	49	58	153	355	65	20	21	338	18	26	1224	1.52	12.44	0.11	1.02	0.32
6	3	7	19	105	22	19	169	514	31	12	36	753	44	23	584	2.46	11.43	0.07	1.01	0.19
3	7	10	36	231	52	70	346	963	163	35	105	1405	95	44	954	4.44	25.80	0.20	2.10	0.47
5	7	7	17	138	42	33	165	295	48	8	18	436	13	15	755	1.70	9.34	0.03	0.84	0.25
10	**10**	**4**	**31**	**148**	**59**	**68**	**304**	**389**	**522**	**33**	**17**	**366**	**17**	**41**	**826**	**1.60**	**16.25**	**0.22**	**5.65**	**0.42**
1	13	4	28	73	26	37	124	180	208	22	14	248	12	16	620	0.91	6.87	0.09	1.58	0.19
7	16	5	23	87	27	31	156	270	86	19	27	354	32	18	406	1.70	8.35	0.06	0.94	0.14
4	24	5	18	98	18	26	129	554	101	14	65	1092	36	19	514	2.96	9.10	0.06	1.26	0.21
11	29	5	23	96	40	32	166	286	59	13	17	290	17	27	587	1.49	10.40	0.07	1.02	0.20
13	43	4	17	81	27	26	154	381	77	21	28	545	28	19	407	1.98	11.02	0.09	1.15	0.16
9	51	4	17	91	28	30	127	241	90	19	14	212	12	20	559	1.26	9.08	0.07	1.37	0.22
14	55	6	34	151	52	43	140	280	134	21	19	391	21	21	1045	1.41	10.44	0.12	1.65	0.30
13	55	8	32	147	72	72	315	690	81	29	47	1140	67	18	1249	2.87	20.52	0.13	1.77	0.44
12	**64**	**6**	**35**	**179**	**45**	**61**	**283**	**588**	**601**	**40**	**49**	**1042**	**76**	**41**	**725**	**3.00**	**15.08**	**0.24**	**6.67**	**0.32**
16	66	2	11	88	76	80	163	302	227	16	7	111	2	30	375	1.01	15.10	0.22	2.79	0.10
Wet zone average		5.8	24.5	125.2	42.3	45.7	192.9	419.2	166.2	21.5	32.3	581.5	32.7	25.2	722.0	2.02	12.75	0.12	2.05	0.26
Intermediate zone																				
15	**67**	**6**	**27**	**55**	**26**	**24**	**113**	**84**	**2713**	**20**	**6**	**175**	**5**	**31**	**4178**	**0.42**	**4.29**	**0.09**	**49.44**	**0.26**
17	72	3	18	74	33	33	112	189	371	17	8	136	7	20	480	0.96	7.56	0.11	2.84	0.17
18	73	5	27	164	94	60	149	333	182	30	12	145	10	33	969	1.12	15.61	0.25	1.58	0.26
20	84	3	23	61	19	18	94	169	331	23	16	226	8	16	720	1.09	5.70	0.06	3.86	0.18
19	**84**	**7**	**33**	**150**	**56**	**56**	**147**	**281**	**91**	**31**	**11**	**186**	**11**	**26**	**1322**	**0.84**	**15.08**	**0.25**	**1.23**	**0.22**
21	86	4	39	151	87	110	224	303	477	51	17	320	12	41	1777	1.40	13.83	0.17	11.04	0.41
22	**91**	**3**	**15**	**69**	**13**	**17**	**121**	**287**	**102**	**20**	**25**	**462**	**37**	**14**	**318**	**1.85**	**8.26**	**0.09**	**1.71**	**0.12**
23	**102**	**3**	**19**	**59**	**18**	**23**	**112**	**208**	**207**	**23**	**14**	**301**	**14**	**13**	**323**	**1.20**	**6.90**	**0.08**	**2.64**	**0.17**
45	**107**	**6**	**34**	**183**	**35**	**32**	**193**	**698**	**323**	**69**	**105**	**1244**	**66**	**39**	**806**	**3.92**	**20.88**	**0.28**	**4.84**	**0.43**
25	**109**	**6**	**39**	**121**	**25**	**39**	**260**	**559**	**444**	**47**	**69**	**916**	**49**	**28**	**578**	**2.97**	**14.26**	**0.15**	**4.25**	**0.38**
26	110	2	17	50	16	18	97	178	175	22	23	191	11	15	363	1.02	8.10	0.11	2.08	0.18
27	**119**	**5**	**43**	**167**	**46**	**62**	**189**	**376**	**595**	**54**	**48**	**538**	**24**	**33**	**929**	**1.70**	**16.33**	**0.22**	**4.87**	**0.50**
28	124	3	13	77	17	17	139	339	119	25	39	412	25	20	273	2.01	10.55	0.12	2.00	0.16
29	**127**	**4**	**27**	**101**	**52**	**59**	**192**	**196**	**273**	**25**	**9**	**170**	**8**	**20**	**714**	**0.69**	**9.56**	**0.10**	**1.80**	**0.19**
30	**131**	**6**	**37**	**87**	**29**	**65**	**217**	**287**	**600**	**36**	**28**	**618**	**54**	**29**	**631**	**1.89**	**10.34**	**0.14**	**5.88**	**0.20**
31	139	4	22	95	43	46	150	232	186	26	18	331	19	20	671	1.06	9.89	0.13	1.64	0.22
32	143	3	17	58	16	22	126	148	167	20	13	270	9	14	330	0.87	6.36	0.07	1.85	0.13
33	151	2	11	34	7	14	102	113	76	14	9	154	6	12	266	0.65	5.46	0.07	1.39	0.09
34	159	1	12	44	8	10	100	105	78	15	9	159	7	11	248	0.68	5.54	0.07	1.42	0.09
35	167	2	10	49	6	17	119	147	70	17	11	170	5	16	241	0.97	6.67	0.09	1.61	0.10
36	**170**	**7**	**32**	**73**	**16**	**17**	**120**	**313**	**828**	**40**	**42**	**1090**	**37**	**16**	**682**	**2.64**	**8.22**	**0.17**	**5.16**	**0.24**
37	173	2	16	34	8	15	79	63	141	13	5	109	3	8	240	0.33	3.75	0.05	1.48	0.08
38	174	1	13	22	4	16	74	45	78	9	3	124	1	5	238	0.24	3.26	0.04	1.14	0.07
39	178	1	10	30	6	10	78	66	54	11	6	127	4	8	236	0.50	4.02	0.05	1.12	0.07
Intermediate zone average		3.7	23.1	83.7	28.3	33.3	137.8	238.3	361.7	27.4	22.8	357.3	18.0	20.3	730.5	1.29	9.18	0.12	4.87	0.21
Dry zone																				
41	182	3	23	93	49	56	133	185	132	23	7	134	7	23	757	0.64	10.15	0.19	1.39	0.21
40	**184**	**8**	**32**	**156**	**29**	**41**	**209**	**762**	**216**	**44**	**80**	**1426**	**141**	**26**	**1065**	**3.54**	**17.41**	**0.18**	**3.07**	**0.25**
42	186	2	10	18	6	11	62	35	83	9	3	112	2	6	258	0.17	2.89	0.05	1.04	0.07
43	205	4	24	91	41	54	142	230	219	27	16	336	16	25	990	1.01	10.53	0.20	1.60	0.19
45	220	2	13	50	6	12	92	214	141	17	25	356	24	12	243	1.8	5.82	0.07	1.65	0.09
44	**229**	**6**	**30**	**166**	**11**	**17**	**263**	**1158**	**379**	**46**	**125**	**1299**	**95**	**31**	**667**	**4.32**	**15.82**	**0.21**	**4.23**	**0.22**
46	245	4	23	51	17	25	96	135	333	22	11	231	8	16	319	0.84	5.31	0.07	2.34	0.17
47	278	4	29	85	13	37	139	353	371	33	42	670	33	14	526	2.07	8.81	0.14	3.51	0.19
50	300	7	35	61	13	26	128	221	577	35	32	531	24	20	16,404	1.69	6.01	0.08	4.12	0.12
49	302	5	39	61	8	31	141	241	526	34	29	535	19	16	2756	1.67	6.17	0.09	4.20	0.15
48	304	6	35	80	28	42	137	213	455	35	24	480	23	18	3635	1.39	8.84	0.13	3.23	0.24
51	306	10	36	63	12	26	169	185	620	30	27	589	31	24	2981	1.61	5.90	0.07	11.34	0.20
52	310	3	25	1114	98	48	270	25	152	14	3	174	4	4	1844	0.27	1.56	0.02	2.42	0.09
53	313	8	29	61	20	25	78	129	2695	31	13	522	18	41	7973	1.06	4.04	0.05	53.72	0.34
Dry zone average		5.1	27.4	153.6	25.1	32.2	147.1	291.9	492.8	28.6	31.2	528.2	31.8	19.7	2887.0	1.58	7.80	0.11	6.99	0.18
Overall average		4.7	24.6	113.5	31.6	36.7	156.2	304.6	338.2	26.0	27.7	466.0	25.8	21.6	1277.0	1.58	9.87	0.12	4.59	0.22

5. Discussion

5.1. Provenance indicators

Elements such as Y, Th, Zr, Nb, Ti and Sc are most suited for provenance and tectonic setting determination (Holland, 1978; Bhatia and Crook, 1986) because of their relatively low mobility during sedimentary processes, and their low residence times in seawater (Holland, 1978; Taylor and McLennan, 1985; Cullers, 1988). These elements are transported quantitatively into clastic sediments during weathering and transport, and hence reflect the signature of the parent material (McLennan et al., 1983a). The

Table 1b
Analyses of soils from the Mahaweli River basin. Major elements as oxide wt%, trace elements ppm. Distance: distance downstream (km) from the uppermost sample point.

S-No.	Distance (km)	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5
Wet zone																				
1	3	2	22	22	12	16	29	58	22	6	3	163	1	11	298	0.62	4.02	0.01	0.53	0.07
2	6	8	46	53	24	15	68	168	204	25	8	163	11	13	272	0.46	6.38	0.07	1.73	0.11
3	13	1	12	78	45	22	68	310	143	20	5	168	2	46	511	0.83	12.98	0.25	2.36	0.11
4	17	10	75	57	10	21	25	8	41	41	6	82	3	1	295	0.20	2.36	0.02	0.57	0.14
5	18	4	25	52	96	100	168	311	2	22	13	153	13	30	339	1.08	13.54	0.16	0.54	0.08
6	18	4	24	94	44	79	154	183	224	24	10	198	12	20	675	0.66	11	0.12	1.08	0.40
7	37	5	8	30	49	35	62	64	44	11	4	59	6	21	2472	0.41	3.45	0.04	18.27	0.14
8	37	1	8	71	56	66	528	284	92	19	6	149	3	42	463	0.72	12.43	0.21	3.83	0.14
9	54	6	12	33	26	23	112	156	4	5	4	159	9	24	616	0.54	7.42	0.04	0.58	0.15
10	72	5	30	89	37	63	103	158	170	30	11	147	31	16	770	0.50	9.28	0.11	1.16	0.24
Wet zone average		4.5	26.3	57.6	39.9	43.9	131.8	169.8	94.6	20.3	6.9	144.0	9.1	22.4	671.1	0.60	8.26	0.10	3.07	0.16
Intermediate zone																				
11	84	4	30	118	60	43	107	178	156	35	11	154	19	23	1118	0.69	9.92	0.07	1.31	0.11
12	89	3	13	65	97	83	720	353	22	18	7	138	8	50	460	0.95	16.85	0.11	1.00	0.10
13	96	5	43	46	26	24	32	83	187	40	4	90	6	1	244	0.27	2.06	0.01	0.60	0.05
14	143	5	21	84	30	39	140	288	262	28	28	643	33	20	873	1.47	9.80	0.21	1.96	0.18
15	154	2	16	33	14	19	87	162	211	13	8	321	12	12	442	1.06	4.56	0.10	1.77	0.07
16	154	2	14	46	8	28	100	277	139	16	16	496	66	11	307	1.98	6.48	0.16	1.52	0.10
17	157	2	13	28	11	20	59	129	191	13	8	249	8	9	366	0.92	4.41	0.09	1.70	0.08
Intermediate zone average		3.3	21.3	60.0	35.1	36.6	177.6	209.8	166.7	23.3	11.8	298.5	21.7	18.1	544.3	1.05	7.73	0.11	1.41	0.10
Dry zone																				
18	161	2	13	47	12	15	65	173	346	15	6	200	5	18	416	0.88	5.71	0.07	2.31	0.09
19	223	1	11	64	13	9	62	215	447	13	6	142	3	20	365	1.06	6.65	0.08	3.42	0.18
20	283	4	23	83	49	61	129	267	212	25	13	297	13	25	609	1.19	9.85	0.13	1.94	0.20
21	289	1	15	27	6	11	78	142	215	13	10	345	3	9	439	1.18	4.33	0.04	2.16	0.08
Dry zone average		2.1	15.4	55.2	20.0	24.0	83.3	199.0	305.0	16.3	8.6	245.9	5.9	18.2	457.3	1.08	6.64	0.08	2.46	0.14
Overall average		3.7	22.6	58.0	34.8	37.9	139.3	188.8	156.3	20.6	8.9	215.5	12.9	20.1	589.8	0.84	7.79	0.10	2.38	0.13

incompatible element pairs Th–Y, Th–Zr, and Th–Nb (Fig. 5a–c) show the effect of heavy mineral concentration and felsic source. Thorium, Y, Zr and Nb abundances in source rocks will increase as their chemistry becomes more evolved. The Th–Y, Th–Zr, and Th–Nb values for the basement rocks show considerable scatter due to the variety of lithotypes analyzed, but most lie within the limits defined by average values in Highland Complex (HC) gneisses (Pohl and Emmermann, 1991). The soils overlap the distribution of the basement rocks, and also lie mainly within the HC gneiss field (Fig. 5a–c). This overlap suggests that the step from basement rock to the soils does not fractionate these ratios in a systematic manner. Although the stream sediments also partially overlap the basement rocks and soils, the data spread to considerably higher values.

The possible role of provenance, sorting or accumulation of heavy minerals such as zircon, monazite, or apatite can be evaluated using Zr/Sc and Th/Sc ratios (McLennan et al., 1993). The Th/Sc ratio is a sensitive index of the bulk composition of the source (Taylor and McLennan, 1985), whereas Zr/Sc ratio serves as a proxy for identifying heavy mineral concentrations, because it is highly sensitive to accumulation of zircon. All elements involved in the ratios are also resistant to weathering processes (Taylor and McLennan, 1985; McLennan et al., 1993). Consequently, plot positions and trends on bivariant Zr/Sc–Th/Sc plots give an indication of source composition and heavy mineral concentration when compared with compositions of average volcanic and plutonic rocks.

Th/Sc ratios of the bulk river sediments, soils, and basement rocks average 1.16, 1.01, and 1.56 respectively, equal to or slightly greater than the ratio in PAAS (0.97, Taylor and McLennan, 1985). The Th/Sc ratio for post-Archean rocks is typically $\sim$1, and for granitic rocks is higher still; for Archean and basic rocks the ratio is less than 1 (Taylor and McLennan, 1985; Fig. 6a). The average Zr/Sc ratio of the sediments is 22.80, somewhat greater than PAAS (17.2; Taylor and McLennan, 1985). This suggests that the sediments are slightly enriched in zircon. The Zr/Sc–Th/Sc plot (Fig. 6a) illustrates that the basement rocks show a range from mafic through to felsic compositions, as do the soils. Scatter among the rocks and soils are considerable owing to the variety of lithotypes analyzed, but most samples fall along a model source evolution line between an average basalt and rhyolite. Basement samples plotting at higher ratios include quartzites and quartzofeldspathic gneisses; higher ratios in these samples may reflect original zircon concentration in their low-grade protoliths. Ratios in the stream sediments show a more restricted range, reflecting homogenization and possibly increased maturity during transport. Zr/Sc and Th/Sc ratios are higher in some samples, suggesting limited zircon concentration. However, these two ratios alone are not adequate to describe provenance, and other combinations of elements must be considered.

Chromium and Ni are often enriched in sediments due to adsorption on clay minerals. Both elements behave similarly in the Mahaweli stream sediments, with broad decrease downstream through the wet and intermediate zones, and gradual increase in the dry zone, suggesting progressive destruction of Cr- and Ni-bearing detrital phases and increased adsorption on to clay minerals. Garver et al. (1996) showed that shale samples derived from ultramafic rocks had high concentrations of Cr and Ni with Cr/Ni ratio of 1.4, suggesting only minor geochemical partitioning from their ultramafic source (Cr/Ni 1.6), whereas sandstones had Cr/Ni ratios of >3.0, showing significant sedimentary fractionation due to concentration of detrital chromite in the sands. In the Mahaweli River sediments Ni is depleted relative to PAAS, whereas Cr is highly enriched, but in the rock samples both elements are depleted (Fig. 4). This may be due to some enrichment process during weathering or sedimentation (McLennan et al., 1983b). The Mahaweli rock samples have average Cr/Ni ratio of 8.9 (including marbles) or 6.1 (excluding marbles), compared to 3.72 for the soils and 5.09 for the river sediments; the PAAS value is 2. The high

Table 1c
Analyses of basement rocks from the Mahaweli River basin. Major elements as oxide wt%, trace elements ppm. Distance: distance downstream (km) from the uppermost sample point.

S-No.	Distance (km)	Rock type	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5
Wet zone																					
1	3	Charnokite	5	38	24	5	3	17	5	175	22	5	132	39	3	306	0.11	1.55	0.01	1.80	0.06
2	4	Leptynite	1	10	3	8	18	54	5	10	4	3	108	3	2	248	0.02	0.05	0.02	0.54	0.03
3	5	Granitic gneiss	4	30	86	11	3	14	28	203	50	19	263	16	6	748	0.44	5.67	0.06	2.10	0.17
4a	6	Garnetiferrous HBG	3	20	63	4	16	55	44	186	27	10	222	30	4	349	0.57	4.60	0.04	2.82	0.25
4b	6	Garnet biotite gneiss	3	34	13	1	4	6	5	135	25	3	78	9	2	248	0.09	1.12	0.01	1.32	0.07
5	6	Marble	0	7	2	0	5	5	5	22	2	2	10	1	22	235	0.02	0.05	0.01	43.37	0.02
6	6	Charnokite	2	11	107	3	5	4	18	24	35	18	657	0	2	367	0.38	6.13	0.14	1.84	0.05
7	14	Charnokitic gneiss	1	6	108	336	42	57	668	107	20	7	87	1	44	4874	2.06	15.79	0.17	8.21	0.26
8	15	Metabasite	1	11	141	7	78	635	284	73	31	8	48	1	50	404	0.68	11.50	0.15	7.71	0.07
9	17	Hornblende biotite gneiss	2	12	95	44	58	108	106	259	18	8	98	5	18	9619	0.56	7.01	0.08	8.70	0.14
10	27	Garnet biotite gneiss	1	10	38	8	85	240	119	422	13	6	97	14	8	283	0.62	4.34	0.03	3.20	0.06
11a	46	Quartzofeldspathc Gneiss	4	36	17	1	5	9	5	119	13	5	320	1	2	251	0.14	0.96	0.02	1.09	0.02
11b	46	Migmatitic gneiss	1	15	39	13	31	39	40	381	11	3	166	1	5	303	0.37	3.36	0.04	3.38	0.07
12	55	Quartzite	0	7	2	3	5	30	5	2	2	1	124	3	2	236	0.06	0.05	0.02	0.55	0.02
13	71	Marble-calk-gneiss	3	9	2	2	0	30	5	60	10	3	52	3	21	745	0.19	0.69	0.02	27.42	0.09
14	71	Marble-blue apatite	2	6	0	2	5	4	5	36	2	2	17	1	19	273	0.02	0.05	0.04	37.23	0.99
15	72	Marble-blue apatite	0	5	2	2	5	5	5	19	2	2	10	0	21	498	0.02	0.05	0.00	40.53	0.03
Wet zone average			2.0	15.7	43.6	26.2	21.6	77.2	79.6	131.3	16.8	6.1	146.4	7.4	13.6	1175.7	0.37	3.70	0.05	11.28	0.14
Intermediate zone																					
16a	80	Hornblende biotite gneiss	3	28	35	1	5	15	5	44	53	14	117	14	2	228	0.12	0.67	0.01	0.84	0.03
16b	80	Granitic gneiss	0	9	78	23	82	87	196	408	17	5	74	1	26	1096	0.97	10.92	0.15	8.47	0.34
17	83	Garnet sillimanite gneiss	7	36	77	3	1	11	6	179	49	19	337	8	1	353	0.40	4.18	0.04	1.77	0.10
18	84	Garnet biotite gneiss	1	8	47	1	60	98	154	59	21	8	127	6	7	245	0.75	6.12	0.08	1.58	0.14
19	85	Charnokitic gneiss	0	6	65	38	5	33	201	201	8	2	58	0	38	1420	0.53	9.95	0.14	9.73	0.09
20	86	Garnet biotite gneiss	0	7	32	1	44	116	134	49	27	7	141	6	9	260	0.75	6.05	0.07	1.45	0.08
21	89	Garnet sillimanite gneiss	1	11	63	2	37	121	140	101	25	8	169	5	14	247	0.75	7.38	0.14	1.20	0.06
22	89	Quartzofeldspathc Gneiss	2	25	29	19	1	25	96	778	32	8	33	14	9	244	0.34	3.93	0.04	2.28	0.47
23	90	Hornblende biotite gneiss	1	11	149	3	59	233	285	81	14	2	56	1	48	318	1.06	11.79	0.16	9.16	0.15
24	94	Garnet biotite gneiss	3	24	54	4	16	60	87	488	16	5	97	3	8	477	0.45	4.06	0.05	3.17	0.22
25	97	Migmatite	2	18	118	75	35	107	175	85	53	17	215	12	21	16,002	1.01	10.90	0.14	2.23	0.04
26	100	Charnockite	1	7	81	29	8	442	241	151	13	4	73	0	42	568	0.67	9.90	0.14	9.50	0.13
27	109	Charnockite	2	19	44	11	5	24	41	336	28	14	209	1	5	386	0.32	4.13	0.06	2.42	0.20
28	155	Charnokitic gneiss	2	12	98	28	14	111	108	1093	17	6	102	2	23	3240	0.52	7.62	0.10	6.24	0.38
Intermediate zone average			1.7	15.6	69.3	17.1	26.5	105.9	133.4	289.5	26.7	8.6	129.1	5.3	18.0	1792	0.62	6.97	0.09	4.29	0.17
Dry zone																					
29	157	Charnokitic gneiss	2	24	15	3	2	14	5	293	13	2	91	1	2	280	0.04	0.58	0.20	1.54	0.05
30	193	Hornblende biotite gneiss	2	9	82	129	137	155	654	212	22	4	120	4	15	14,421	0.53	3.40	0.02	3.30	0.26
31	223	Charnokitic gneiss	1	11	91	8	10	38	125	369	28	10	136	1	23	449	0.78	6.54	0.09	5.04	0.22
32	283	Hornblende biotite gneiss	1	10	56	21	5	14	14	1878	21	6	102	2	18	254	0.25	3.14	0.05	13.66	0.12
33	284	Biotite gneiss	3	21	51	9	7	13	62	1754	18	11	88	21	3	291	0.48	3.28	0.03	3.00	0.36
34	286	Charnokitic gneiss	1	9	66	17	11	37	107	440	26	9	98	3	21	507	0.60	6.97	0.17	5.36	0.24
35	304	Charnokitic gneiss	2	14	85	3	2	6	10	112	22	9	375	1	4	310	0.28	3.77	0.09	1.76	0.09
Dry zone average			1.6	13.9	63.8	27.1	24.7	39.5	139.4	722.6	21.6	7.4	144.3	4.8	12.3	2359	0.42	3.95	0.09	4.81	0.19
Overall average			1.8	15.3	56.7	22.9	24.0	81.3	110.2	294.1	21.4	7.3	139.5	6.1	15.0	1614	0.47	4.97	0.07	7.53	0.16

Table 2
Average, minimum and maximum values for the bulk stream sediments, basement rocks, soils and tributaries of the Mahaweli River basin compared to the PAAS values of Taylor and McLennan (1985). Major elements as oxide wt%, trace elements ppm.

	Sediment			Rock			Soil			Tributaries			PAAS
	Avg	Min	Max	Avg	Min	Max	Avg	Min	Max	Avg	Min	Max	
Fe_2O_3	9.83	1.56	25.80	4.95	0.05	15.79	7.77	2.06	16.85	**20.88**	4.29	12.76	7.18
TiO_2	1.57	0.17	4.44	0.47	0.02	2.06	0.84	0.20	1.98	**4.32**	0.42	2.18	0.99
MnO	0.12	0.02	0.28	0.07	0.00	0.20	0.10	0.01	0.25	0.28	0.08	0.17	0.11
CaO	4.63	0.84	53.72	7.51	0.54	43.37	2.40	0.53	18.27	**49.44**	1.23	7.25	1.29
P_2O_5	0.21	0.07	0.50	0.16	0.02	0.99	0.13	0.05	0.40	0.50	0.12	0.28	0.16
As	5	1	10	2	0	7	4	1	10	8	3	6	5
Pb	25	10	43	15	5	38	23	8	75	43	15	31	20
Zn	114	18	1114	57	0	149	58	22	118	183	55	122	85
Cu	31	4	98	23	0	336	34	6	97	59	11	33	50
Ni	37	10	110	24	0	137	38	9	100	68	17	41	55
Cr	156	62	346	81	4	635	138	25	720	**304**	112	194	110
V	304	25	1158	110	5	668	189	8	353	**1158**	84	442	150
Sr	341	31	2713	299	2	1878	159	2	447	**2713**	91	564	200
Y	26	8	69	21	2	53	21	5	41	**69**	20	38	27
Nb	28	3	125	7	1	19	9	3	28	**125**	6	45	19
Zr	466	109	1426	140	10	657	215	59	643	**1426**	170	702	210
Th	26	1	141	6	0	39	13	1	66	**141**	5	45	15
Sc	22	4	44	15	1	50	20	1	50	41	13	28	16
TS	1298	236	16,404	1621	228	16,002	588	244	2472	**4178**	318	983	621

Bold values for tributary averages indicate high contents.

ratios for the Mahaweli rocks, soils, and sediments thus indicate that Cr has undergone significant fractionation from source rock, to soil, to sediment, and also that no significant ultramafic component is present. Nevertheless, Cr–Ni relations clearly indicate that the Highland Complex samples are post-Archean (2–3 Ga) in nature rather than late or early Archean (Fig. 7a). This figure shows abundances in the basement rocks are highly variable, whereas the river sediments show less scatter but higher Cr contents,

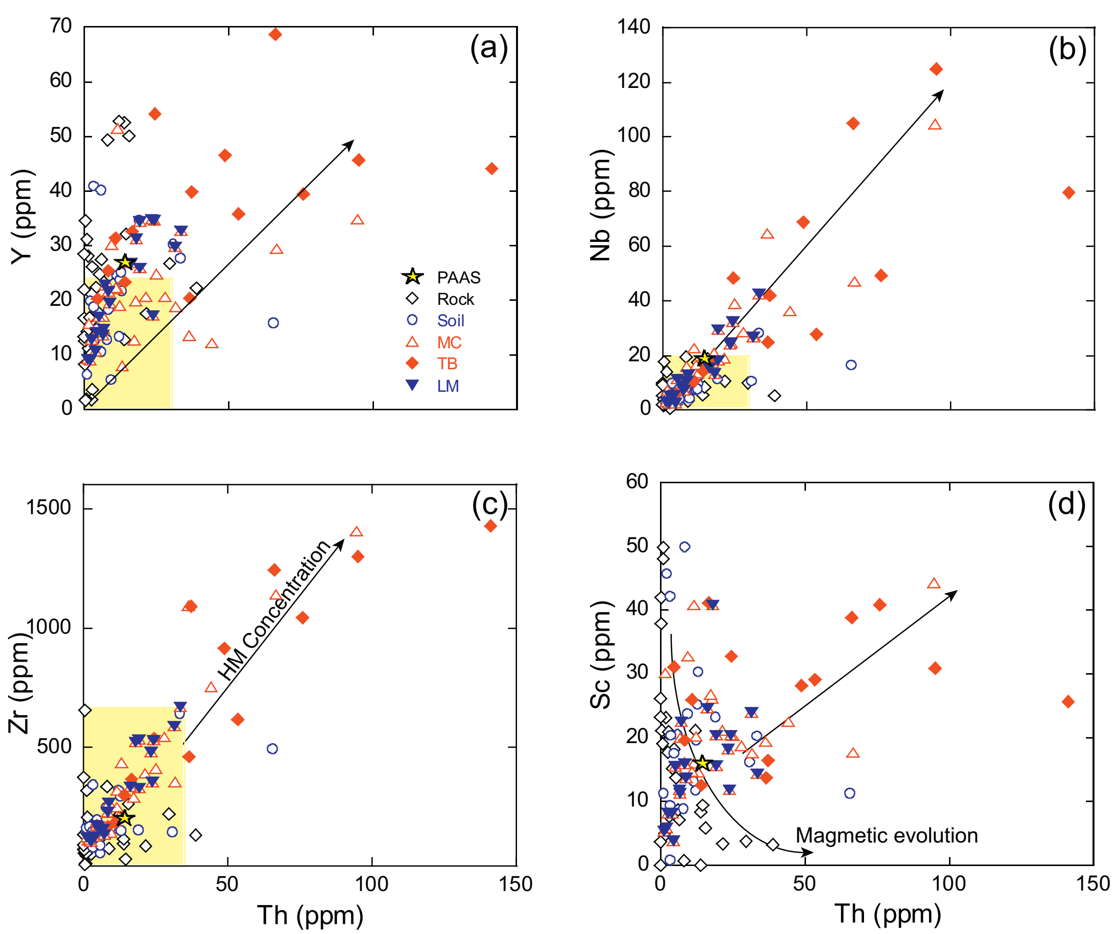

Fig. 5. Plots of (a) Y, (b) Nb, (c) Zr and (d) Sc versus Th for the bulk stream sediments, rocks and soils. Stream sediments are differentiated by location: MC – main channel; TB – tributaries; LM – lower main channel. Yellow areas indicate typical ranges in alkaline rocks in the Highland Complex (data from Pohl and Emmermann, 1991). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

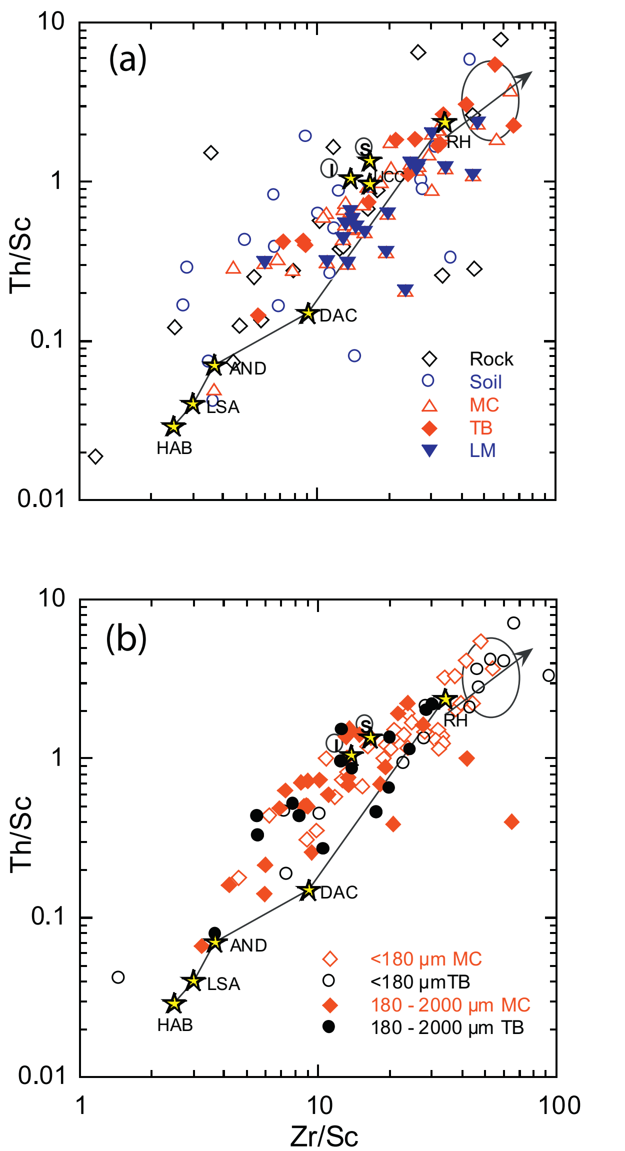

Fig. 6. Zr/Sc–Th/Sc ratio plots (McLennan et al., 1993) for (a) bulk stream sediments, basement rocks and soils from the Mahaweli River, showing zircon concentration (arrow) and typical source rock compositions; (b) <180 μm and 180–2000 μm fractions. Stars BAS, AND, DAC, RHY: average basalt, andesite, dacite and rhyolite, as plotted by Roser and Korsch (1999); stars (I) average I-type granite, (S) average S-type granite (Whalen et al., 1987).

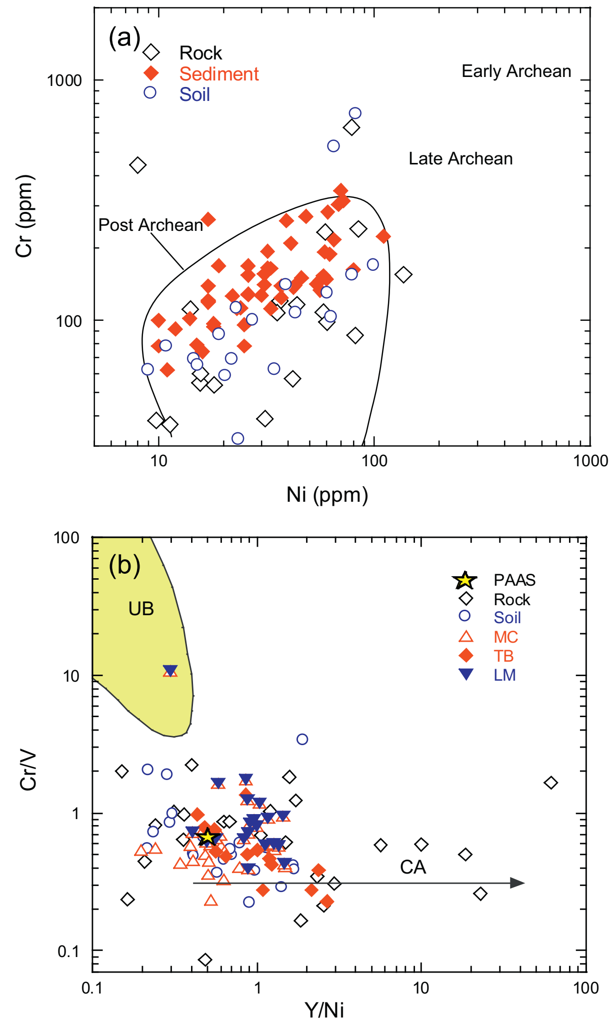

Fig. 7. (a) Cr–Ni plot for the Mahaweli stream sediments, rocks and soils, showing the post-Archean field (Taylor and McLennan, 1985) and fractionation from source rocks to the sediments. (b and c) Cr/V–Y/Ni plots (McLennan et al., 1993) for the stream sediments, rocks and soils (b); and <180 μm and 180–2000 μm fractions (c), showing the lack of ultrabasic sources. UB: ultrabasic; field of sands derived from ultrabasic rocks in the Hino River, South West Japan (Ortiz and Roser, 2006b); CA: typical calc-alkaline trend.

indicating homogenization and enrichment during weathering and transport.

Cr/V–Y/Ni ratios also provide estimates of preferential concentration of chromium over other ferromagnesian elements (Hiscott, 1984; McLennan et al., 1993). The Cr/V ratio measures enrichment of Cr with respect to other ferromagnesian elements, whereas the Y/Ni ratio evaluates the relationship between the ferromagnesian trace elements (represented by Ni) and the HREE, using Y as a proxy (McLennan et al., 1993). Sediments derived from ultrabasic sources typically have high Cr/V ratios (≫1) coupled with low Y/Ni (<1) (Hiscott, 1984), as shown by Ortiz and Roser (2006b) for sediments from the Hino River in SW Japan. Very high Y/Ni ratios are seen in some basement rock samples (Table 1c), such as Digana-garnet sillimanite gneiss (61.7), Gurudeniya marble (34.3), Kandehandiya quartzo-feldspathic gneiss (22.9), Ginigathhena granitic gneiss (18.5), Victoria hornblende biotite gneiss (11.1) and Girandurukotte – charnockitic gneiss (8.9). Average Cr/V ratios of the rocks, soils and river sediments are all low (0.80, 0.79, 0.84 respectively), whereas Y/Ni ratios (average 0.87, 0.76 and 6.41) are variable. Cr/V ratios in both the soils and river sediments are low, indicating that contribution from ultrabasic sources is negligible (Fig. 7b). Y/Ni ratios generally range across values typical of intermediate to felsic calc-alkaline rocks.

5.2. Sorting

In stream sediments Fe_2O_3 and TiO_2 can be expected to be positively correlated due to sorting effects (Singh, 2009). Linear arrays of data points along lines extending toward the origin in binary Fe_2O_3–TiO_2 plots demonstrate that the elements were immobile, and that they were also hydraulically fractionated in a similar manner. A Fe_2O_3–TiO_2 plot for the Mahaweli data (Fig. 8a) shows the basement rocks form a linear trend with TiO_2/Fe_2O_3 ratios of ~0.086, similar to the ratio in PAAS (0.099; Taylor and McLennan, 1985). While many of the soils plot within the field of the basement rocks, some also scatter to higher TiO_2/Fe_2O_3. TiO_2 can accumulate in residual soils due to loss of more mobile constituents (Gracia et al., 1994; Young and Nesbitt, 1998), and hence may reflect degree

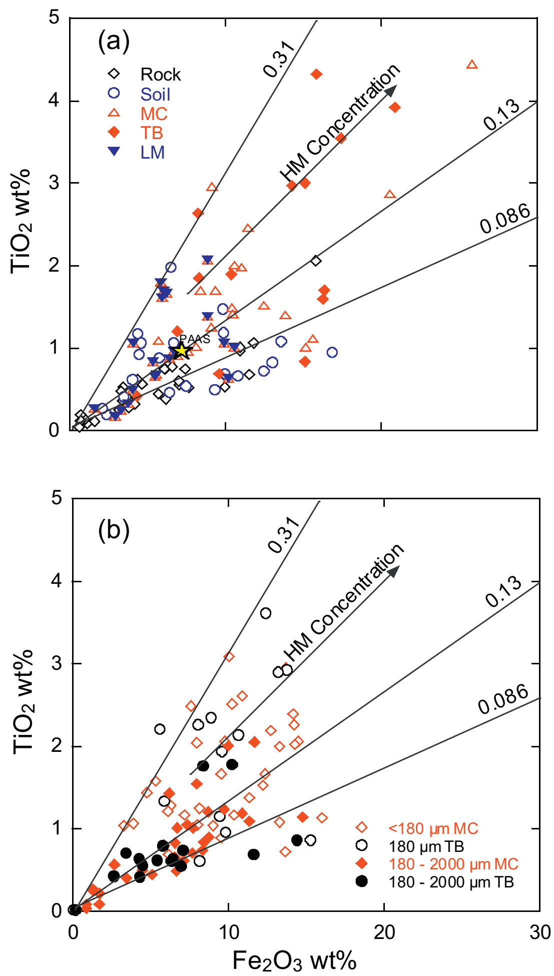

Fig. 8. Fe_2O_3–TiO_2 plots for (a) bulk stream sediments, rocks and soils, and (b) the <180 μm and 180–2000 μm fractions, illustrating hydraulic fractionation and heavy mineral concentration. MC – main channel; TB – tributaries; LM – lower main channel.

of weathering. However, examination of the soil data by climatic zone shows there is no consistent association of higher TiO_2 abundances with the soils in the wet zone, and hence weathering does not seem to control TiO_2 content. Most Mahaweli River sediments have TiO_2/Fe_2O_3 ratios similar to the soils, and trend toward the origin, suggesting hydraulic fractionation and quartz dilution (Singh, 2009). As with the soils, some stream sediments have much higher TiO_2 contents, coupled with high Fe_2O_3, with TiO_2/Fe_2O_3 ratio between 0.13 and 0.31 (Fig. 8a). This suggests concentration in the sediments of a heavy mineral phase containing both Ti and Fe, such as ilmenite and magnetite.

Sorting effects are also evident when the compositions of the stream sediment size fractions. Fe_2O_3–TiO_2 ratios for the 180–2000 μm fractions in both the main channel (MC) and tributaries (TB) fall between 0.086 and 0.13, whereas ratios in the <180 μm fractions are mainly between 0.13 and 0.31 (Fig. 8b). However, most of the <180 μm fractions in the tributaries have much higher Fe_2O_3 values than their 180–2000 μm fractions. This pattern is also seen in the main channel. The size fraction data thus confirm that hydraulic fractionation of Fe- and Ti-bearing phases has occurred.

A Th/Sc–Zr/Sc plot (Fig. 6b) for the fractions shows higher values in <180 μm TB samples than in the MC <180 μm equivalents. The MC and TB 180–2000 μm fractions exhibit similar Th/Sc–Zr/Sc values to each other. On the other hand, the TB and MC <180 μm

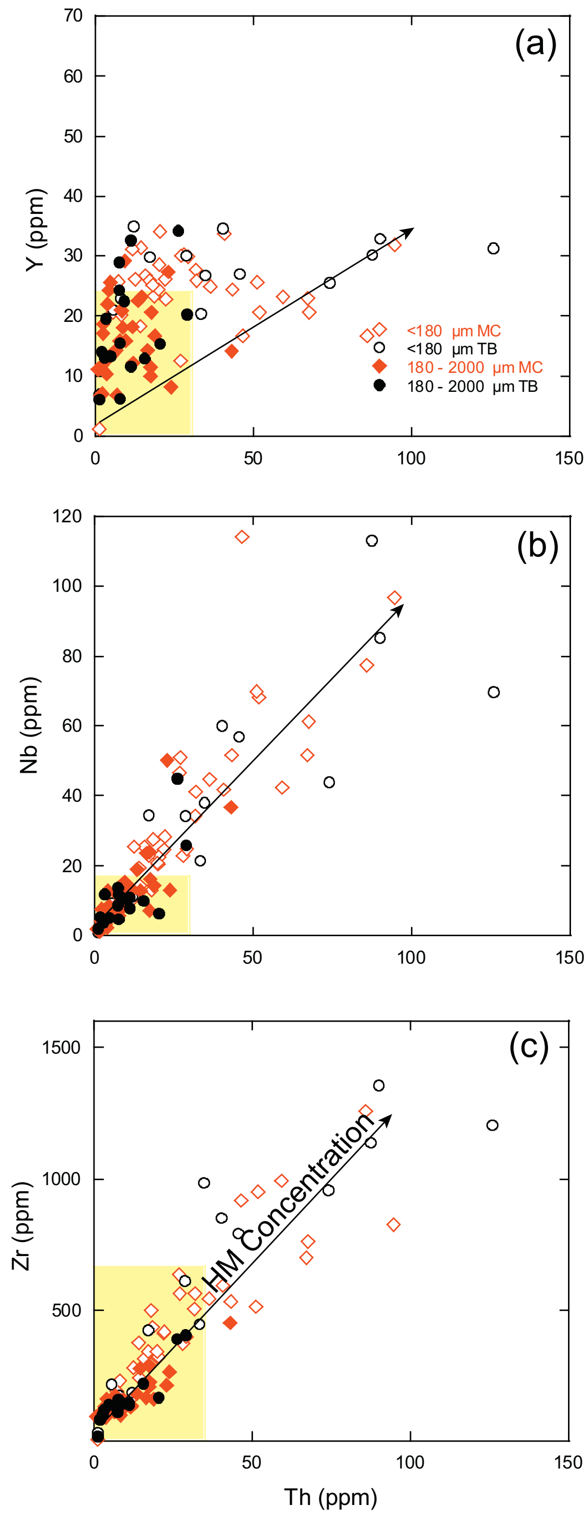

Fig. 9. Plots of (a) Y, (b) Nb, and (c) Zr versus Th for the <180 μm and 180–2000 μm fractions. MC – main channel; TB – tributaries. Yellow areas indicate typical ranges in alkaline rocks in the Highland Complex (data from Pohl and Emmermann, 1991). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

fractions both show higher values than their 180–2000 μm equivalents (Fig. 6b). This indicates Zr enrichment in the fine fractions in both the tributaries and the main channel, suggesting transport distance does not greatly affect sorting. Moreover, average Th/Sc (1.01) and Zr/Sc (15.29) ratios in the MC 180–2000 μm fractions are low, compared to average values of 3.31 and 25.16 respectively in the <180 μm fraction. These results show that both Th and Zr are enriched in the fine fractions of both the MC and TB samples as a result of sorting.

5.3. Heavy mineral indicators

As noted above, elements such as Y, Th, Zr, Nb, Ti and Sc are well suited for provenance and tectonic setting determination (Holland, 1978; Bhatia and Crook, 1986), but they are also good indicators of heavy minerals. Due to high durability, these minerals tend to remain in sediments during transport over long distances. Tourmaline, zircon, and rutile are associated with significant enrichments of Th, Zr and Nb, and suggest a granitic source or recycled detritus. The sediment fractions are useful for detailed description of the heavy minerals related to Th, Zr, Nb and Y.

Niobium and Th contents of the Mahaweli River rock and soil samples are lower than those in the stream sediments, in which the maximum Nb content reaches 125 ppm (Table 2). There is also no close association between the Nb values of basement rocks and their adjacent sediments. Zirconium shows similar variation to Th, and is strongly enriched in the river sediments relative to the soils and basement rocks (Table 2). The river sediments also show very strong internal correlations between Zr and Nb (+0.91), Zr and Th (+0.91), and Nb and Th (+0.86).

Distributions of these elements in the stream sediments are likely to be controlled by heavy minerals including zircon, monazite, garnet and titanite, with the lesser variability being produced by homogenization of the detrital bedload during transport. For Zr the scatter to higher values is relatively coherent, suggesting zircon concentration is the main cause of the enrichment. The spread to higher values is more scattered for Y and Nb, suggesting that multiple heavy minerals control their abundances. In contrast, Sc abundances in the basement rocks range up to 50 ppm, but decrease with increasing Th, as expected during magmatic evolution (Fig. 5d). Soils show similar values and considerable scatter, but some stream sediments scatter to higher values of both Sc and Th, suggestive of different heavy mineral concentration in these samples.

The incompatible element pairs Th–Y, Th–Zr, and Th–Nb (Fig. 9) for the <180 μm and 180–2000 μm fractions of the sediments show higher abundances of Y, Nb and Zr are present in the <180 μm fractions. The tributaries also show high values in the <180 μm fraction, suggesting that preferential concentration of heavy minerals in the fine fraction is not a function of transport distance.

5.4. Climatic zone variation and weathering

Weathering can be determined using various methods. However, most of the commonly used weathering indices such as CIA (Nesbitt and Young, 1982), PIA (Fedo et al., 1995), CIW (Harnois, 1988), and WIP (Ohta and Arai, 2007) include CaO in the calculations. The carbonate rocks in the Mahaweli River catchment naturally have high Ca contents, and LOI values of the soils and sediments range between 0.61–12.86 and 0.31–16.18 wt%, respectively. Due to the very high carbonate contents, the CIA, PIA, CIW, and WIP indexes cannot be used for weathering interpretations in the Mahaweli River. However, the ratios of immobile to mobile elements can be adopted as a measure of the degree of weathering and mobility (Nesbitt and Young, 1982; Gallet et al., 1996). In this study K_2O/Na_2O and $Al_2O_3/(K_2O + Na_2O)$ ratios (Fig. 10a and b) of the bulk stream sediments have been used to describe the weathering and elemental mobility along the course of the river across the three climatic zones. The $Al_2O_3/(K_2O + Na_2O)$ ratios (Fig. 10a) of the sediments show a clear change across the climatic zones. The wet zone sediments are Al-rich indicating the presence of a considerable amount of clay derived from feldspars, and intense weathering in the elevated parts of the upper catchment, with decreasing intensity downstream. The lack of fine-grained material in the stream sediments suggests that mechanical weathering has also occurred. However, the presence of a considerable amount of Na (2.14 wt%; Young and Ishiga, unpubl. data) in the wet zone stream sediments indicates that the weathering process is not complete. $Al_2O_3/(K_2O + Na_2O)$ ratios decline across the intermediate zone, and level out in the dry zone, despite the quartzose and well-sorted nature of the sediments in the lower reaches of the river.

K_2O/Na_2O ratios can be strongly modified by both diagenesis and weathering. However, K_2O/Na_2O ratios show no clear trend with the climatic zones along the Mahaweli River (Fig. 10b). High K_2O/Na_2O ratios at some elevated sites with granites and metabasites in the wet zone indicate intense weathering (Fig. 10b). However, moderately high contents of K_2O at some of the wet zone stream sediment locations suggest that it is incorporated mainly in felsic fragments, because contents of potassium-rich minerals like K-feldspars, muscovite and illite are generally low in the sand fractions. The cause of the variable Na_2O concentrations between climatic zones is not yet clear; the variability may be related to both source weathering and plagioclase (albite) contributions from tributaries.

The extent and type of vegetation has a complex effect on chemical weathering rates (Stallard, 1992). The upper reaches of the Mahaweli catchment area are heavily vegetated. This may affect weathering rates in a complex manner, by decreasing the exposure of fresh bedrock, while increasing moisture retention in the upper catchment. However, the type and extent of vegetation varies with precipitation and temperature, and any effect from vegetation may be incorporated in the overall weathering regime in each climatic zone.

Soils are the main products of weathering. LOI (Loss On Ignition) values for the soils in the Mahaweli River catchment (Fig. 10c) can be used as a rough measure of the weathering conditions at each site. High CaO contents of the soils (Table 1b) indicate the presence of carbonates, because gypsum cannot have formed, based on the low TS contents. Organic matter is another factor that could be associated with high LOI. The LOI values of the soils (Fig. 10c) correlate well with the $Al_2O_3/(K_2O + Na_2O)$ weathering profile (Fig. 10a), and show the same trend down the river. The high LOI values in the wet zone soils may partially controlled by the organic matter produced by vegetation in that area. The upper part of the Mahaweli catchment is covered by dense forest, and tea and vegetable cultivation, combined with very high rainfall. The forest almost disappears toward the dry zone, where the vegetation is mainly paddy. The dense vegetation cover in the wet zone and chemical weathering has thus combined to increase the LOI in the soils.

The alkaline earth elements Sr and Ca can also be used as weathering indicators. Strontium is depleted in sediments during weathering of feldspar and with increasing sediment maturity (Bhatia and Crook, 1986). The Mahaweli River sediments show strong Sr depletion in the wet zone, where intense weathering of feldspar can be anticipated (Fig. 11). Abundances in the stream sediments increase sharply in the intermediate zone with the entry of four major tributaries (Katugasthota, Raththota-Sudugaga, Huluganga, Maoya) which contain marbles in their catchments. Values are higher in the Katugasthota and Raththota-Sudugaga than in the Huluganga and Maoya, after which the values drop. The Katugasthota and Rattota-Sudugaga catchments have higher elevations and receive more rainfall than the Huluganga and Maoya catchments. The Katugasthota and Raththota-Sudugaga are thus much larger rivers and carry more water and sediment to the main river. Intense mechanical weathering due to high rainfall and erosion are thus likely to be responsible for the spikes in the

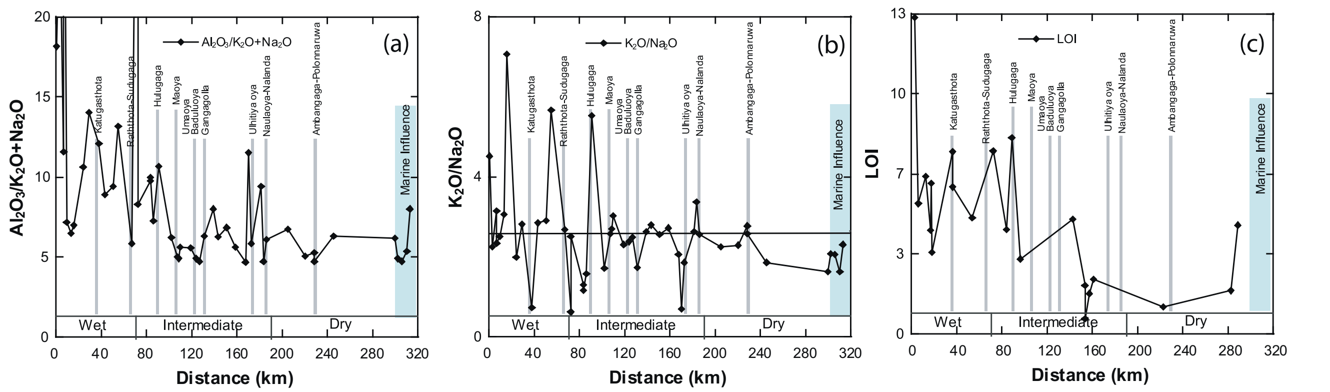

Fig. 10. (a) $Al_2O_3/K_2O + Na_2O$ (b) K_2O/Na_2O ratios in bulk sediments down the Mahaweli River showing the weathering trend with the climatic zones (Young and Ishiga, unpublished data). (c) Loss On Ignition of soils down the Mahaweli River.

Katugasthota and Raththota-Sudugaga catchments. Strontium contents then decrease steadily through the intermediate zone, suggesting progressive weathering of calcareous detritus during transport. Abundances increase slightly downstream in the dry zone, probably due to contribution of detritus from local calc-silicate rocks, before increasing abruptly in the lower reaches and Trincomalee Bay due to marine influence. A similar pattern is observed for CaO (Fig. 11). Strontium and CaO contents of soils in the wet zone also tend to be less than those in the intermediate and dry zones (Table 1c). Ranasinghe et al. (2008) observed positive correlation of CaO and Sr in river sediments in the upper Mahaweli, and these elements are also very highly correlated (+0.97) in the sediments in our study. The Sr and CaO contents in the Mahaweli River sediments are thus controlled by both weathering and source rock composition, with major contributions of CaO and Sr from tributaries containing marbles.

A plot of Cr/V ratios in <180 μm and 180–2000 μm fractions along the main channel (Fig. 12a) shows that Cr/V is consistently greater in the 180–2000 μm fraction. Spikes are seen at the tributary confluences. Contrast between the fractions is slightly higher in the wet zone than in the intermediate zone, but is greatest in the dry zone. A plot of the relative enrichment between the fractions (Cr/V_{EF}) downstream (Fig. 12b) clearly shows that Cr is progressively lost from the 180–2000 μm fractions in the wet zone, possibly by oxidation of reactive Cr-bearing phases such as chrome spinel (Dissanayake, 1982), chromite (Dissanayake et al., 2000) and hercynite and spinel (Ranasinghe et al., 2008) hence transferring Cr to the fine fraction. The Cr/V_{EF} then steadily increases through

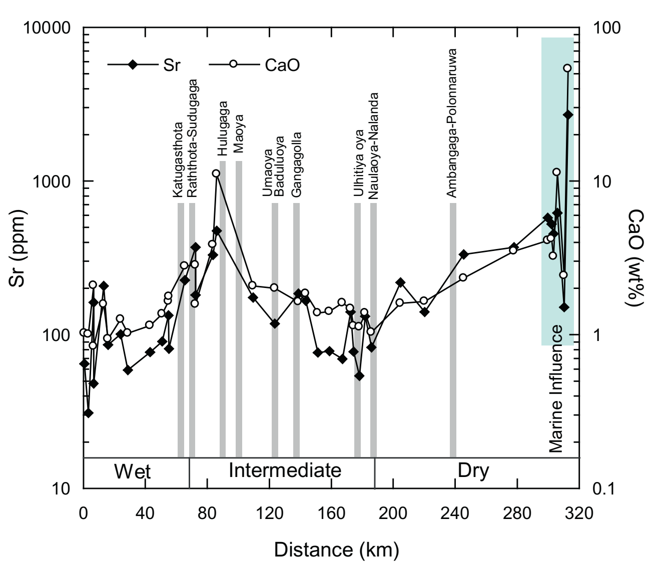

Fig. 11. Variation in CaO and Sr abundances in bulk stream sediments down the Mahaweli River, also showing tributary entries and the climatic zones.

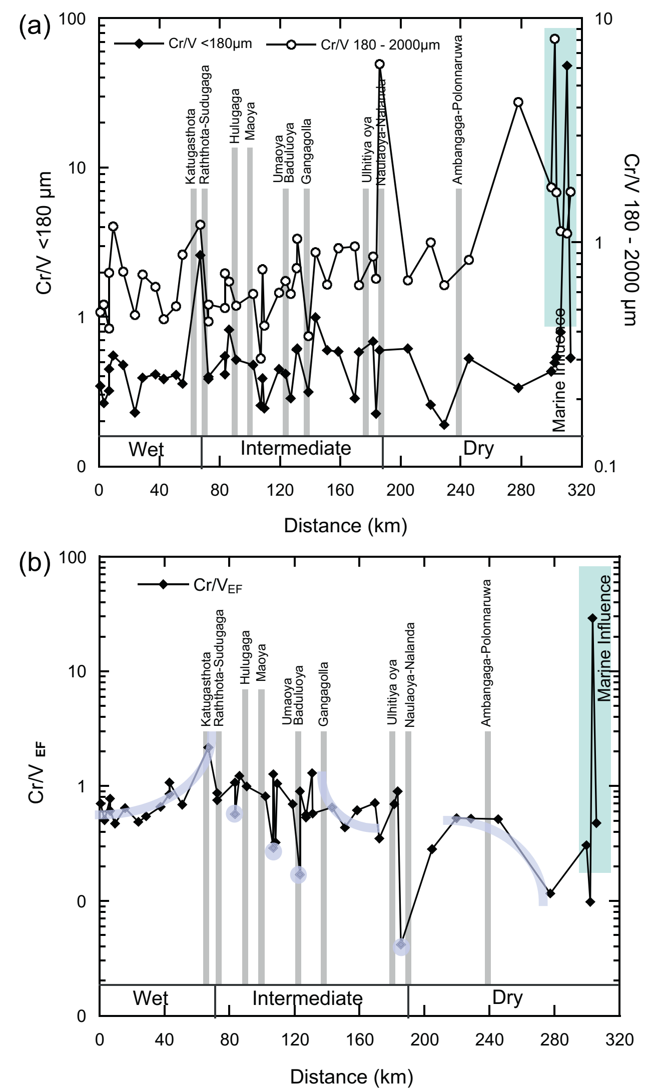

Fig. 12. (a) Variation in Cr/V ratios in <180 μm and Cr 180–2000 μm fractions in sediments down the Mahaweli River, also showing tributary entries and the climatic zones. (b) Cr/V Enrichment Factor (Cr/V_{EF}) downstream, derived by normalizing Cr/V in the 180–2000 μm fraction against Cr/V in the <180 μm fraction.

the intermediate and dry zones, apart from a large spike where the Ulhitiya Oya and the Naula Oya-Nalanda tributaries enter the main channel (Fig. 12b), and a large drop with the marine influence in Trincomalee Bay. This suggests that the intermediate zone tributaries supply relatively fresh coarse-grained Cr-bearing detritus to the main channel, which remains in the 180–2000 μm fraction down to the sea. This pattern contrasts with the findings of Ortiz and Roser (2006a), who observed a steady decline in Cr/V ratio downstream in the Hino River of SW Japan. This was attributed to progressive oxidation of ultramafic detritus and Cr-bearing heavy minerals, moving Cr from the 180–2000 μm fraction to clays in the <180 μm fraction. In the Mahaweli River this trend is only observed in the wet zone (Fig. 12b).

5.5. Tributary effect

Stream sediments in most of the tributaries have high elemental concentrations compared to the soils and rocks (Table 1a, highlighted rows represent tributaries; Table 2). Most of the tributaries rise in elevated areas of the Mahaweli catchment, mainly in the wet and intermediate zones. The source rocks should thus have been subjected to intense weathering. The tributaries display the highest concentrations of Y, Th, Zr, Nb, Ti Sc and Fe (Figs. 6b, 8b and 9a–c) suggesting that the source rocks within them are mainly of mafic composition, and can contribute considerable amounts of heavy minerals. The high values are probably related to the local lithology in the catchment of each tributary. The main lithologies present are garnet-biotite gneiss, hornblende-biotite gneiss, granitic gneiss, charnockite and metabasite. The Umaoya and Baduluoya are the longest of the tributaries, but they do not display high concentrations of the above elements. The Umaoya and Baduluoya thus do not contribute greatly to the load of these elements in the main river channel. In the main channel, most of these elements tend to accumulate downstream (Table 1a). The tributaries contribute to these accumulations mainly at the confluences. However, each tributary contains rocks of differing composition, and their contributions to the elemental loadings in the main river channel also differ as a result (Table 1a). More detailed studies of individual tributaries are required to evaluate local lithological controls, and the contribution of each tributary to the Mahaweli sediment load.

5.6. Environmental effects

The enrichment factor (EF) is a good tool to evaluate potential environmental contamination, as it can be used to differentiate between lithogenic and naturally occurring metal sources (Zhang et al., 2009; Olubunmi and Olorunsola, 2010; Naji and Ismail, 2011) and to assess the degree of anthropogenic influence. EF has here been calculated using Fe as a normalizer, because iron is the fourth most abundant major element in the Earth's crust, and is usually of no contamination concern. The EF for Fe-normalized data is defined by Sutherland (2000) as:

$$\mathrm{EF}_{\mathrm{metal}} = \frac{(M_x/\mathrm{Fe}_x)_{\mathrm{sample}}}{(M_c/\mathrm{Fe}_c)_{\mathrm{PAAS}}};$$

where M_x and Fe_x are the concentrations of the metal and Fe in the sample, respectively, and M_c and Fe_c are the concentrations of the metal and Fe in PAAS, used as an index of upper continental crust composition.

Evaluation of the EF varies a little between authors. Zhang and Liu (2002) state that EF values between 0.5 and 1.5 indicate the metal is derived entirely from crustal materials or natural processes. EF values >1.5 suggest anthropogenic sources. Sutherland (2000) indicated EF values of <1 indicate background concentrations; values of 1–2 depletion to minimal enrichment suggestive of no or minimal pollution; 2–5 moderate enrichment; 5–20 significant enrichment; 20–40 very high enrichment; and >40 extremely high enrichment.

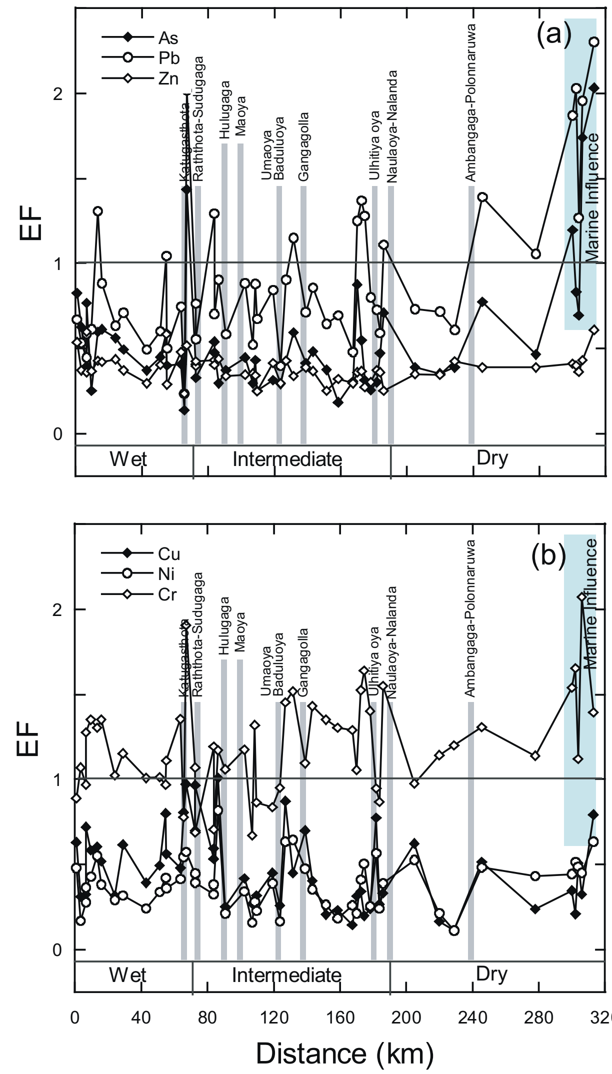

Fig. 13. Fe-normalized enrichment factors for (a) As, Pb, and Zn, and (b) Cu, Ni, and Cr down the Mahaweli River, also showing tributary entries and the climatic zones.

Plots of the EF for As, Pb, Zn, Cu, Ni and Cr along the main channel of the Mahaweli River are shown in Fig. 13. With the exception of Cr throughout the river and Pb at a few sites in the dry zone, all other elements are strongly depleted, with EF < 1. According to the criteria of both Sutherland (2000) and Zhang and Liu (2002), As, Zn, Cu and Ni abundances in the Mahaweli sediments thus represent background values, and there is no threat of anthropogenic contamination. The slight Cr and Pb enrichments are related to the local lithology, since there are no industrial point sources of contamination within the area. Spikes at the entries of tributaries are likely to be caused by heavy minerals, and there is no significant change seen with climatic zone. Furthermore, most individual Cr and Pb EFs are <1.5, within the limit for natural sources proposed by Zhang and Liu (2002).

The single Trincomalee harbor sample analyzed has been removed from these plots, since it has high EFs for all elements. The EFs for As, Pb, Zn, Cu, Ni and Cr in this sample are 2.0, 5.1, 28.9, 10.1, 3.26 and 12.5, respectively. It is thus moderately enriched in Pb and Ni, significantly enriched in Cu and Cr, and very highly enriched in Zn, using the criteria of Sutherland (2000). Consequently, there is potential for significant anthropogenic contamination in Trincomalee harbor. This will be examined in future work.

The only similar study carried out in the Mahaweli River was a detailed survey by Ranasinghe et al. (2008), but this was confined to the upper reaches. Some of the results of our study compare well with Ranasinghe et al. (2008), but others do not. Chromium contents in the stream sediments in our study are lower. Lead and Y abundances are almost the same in both studies, and the areas where anomalous Pb values occur correspond. Nickel, V and Sr show much higher values in our current study. Ranasinghe et al. (2008) also observed low Sr values in the wet zone. Copper and Zn values are lower in our current study. The contrasts seen between these two datasets are likely to be a product of the greater variability and regional extent of our sample collection.

6. Conclusions

Elemental abundances in Mahaweli River sediments suggest some loss of mobile components from the source rocks and soils during weathering in the three climatic zones, and homogenization of the stream sediments during transport. The lack of fine-grained material in the stream sediments suggests that mechanical weathering has also occurred. The presence of considerable Na_2O in the wet zone stream sediments indicates that the weathering process is not complete. The upper reaches of the Mahaweli catchment area are heavily vegetated, and this affects weathering rates in a complex manner. Most of the stream sediments reflect a felsic source overall, whereas the basement rocks show a range of composition from mafic through intermediate to felsic compositions with a variety of lithotypes, as do the soils. Ti and Fe concentrations in the sediments partly reflect hydraulic fractionation and also concentration of heavy mineral phases containing both Ti and Fe, such as ilmenite and magnetite. High concentrations of Y, Th, Zr, Nb, Ti and Sc indicate significant concentration of heavy minerals (zircon, monazite, garnet, titanite, rutile and tourmaline), especially in the <180 μm fractions. Cr/V ratios and Sr and CaO show some variation with climatic zone, illustrating the link between climate and weathering intensity. The tributaries contribute considerably to elemental loads in the main river channel, but more detailed studies of individual tributaries are required to evaluate local lithological controls, and the contribution of each tributary to the Mahaweli sediment load. There is little evidence for contamination in the Mahaweli, with EF values for As, Zn, Cu and Ni all <1, and slightly higher values for Cr and Pb still within background limits. The results overall suggest that the composition of active sediments in the Mahaweli River is mainly influenced by source lithology, climate, weathering, hydraulic sorting and transport.

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