Elemental Composition and Sources of Atmospheric Particulate Matter in Dar es Salaam, Tanzania

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Abstract

An intensive aerosol field campaign was carried out from 16 August to 16 September 2005 (dry season) at a kerbside in Dar es Salaam, Tanzania. A Gent PM_{10} stacked filter unit sampler with coarse and fine Nuclepore polycarbonate filters, providing fine (0.4 µm) and coarse (8 µm) size fractions, was deployed. A total of 64 parallel collections were made. All samples were analysed for the PM mass by weighing. A further analysis was performed for 25 elements by particle-induced x-ray emission spectrometry. The PM_{10} mass, as derived from the stacked filter unit samples, was, on average, 58 µg/m³. The concentrations of the heavy metals were lower than those for the elements of crustal origin. Nevertheless, some typical anthropogenic metals, such as Zn and Pb, exhibited much higher median PM_{10} levels, suggesting strong local sources for these elements in Dar es Salaam. The results also showed very strong day/night differences for the crustal elements (Al, Si, Ca, Ti and Fe). Most elements exhibit strong correlations in the coarse size fraction and somewhat weaker ones in the fine size fraction suggesting that they may originate predominantly from the same source. Principal component analysis with VARIMAX rotation was applied to the data set. Five and four components were identified for the fine and coarse fractions and explained 86.5% and 90.8% of the variance in the data set respectively.

Keywords: PIXE; Atmospheric Aerosols; Elements; Size Fractions; PCA; Kerbside

Introduction

Air pollution has become one of the most environmental concerns in urban areas, especially in view of the adverse health effects associated with ambient atmospheric particles (Pope, 2000). In the urban environment, particulate matter may be generated among others by automotive vehicles (road dust), manufacturing processes, fossil fuel combustion, biomass burning and through photochemical reactions involving precursors gases. The identification of various sources of airborne particulate matter (PM) and the assessment of their impact on the aerosol composition are some of the major goals of contemporary atmospheric research. Among the various species present in the particulate matter, a concern have been on the study of toxic metals such as lead, zinc, copper etc and on elements of great diffusion in the environment (IPCC, 2007). Because of local and global contamination of these elements and their relative effects on human health, a detailed study of the elemental composition of atmospheric aerosols and sources should give invaluable information.

In Tanzania few measurements of PM, its composition and sources are available (Bennet et al., 2005; Mkoma et al., 2009a,b). The objective of this research work was to explore the use of the multi elemental capability of the Particle Induced X-Ray Emission (PIXE) technique for determining element spectra in the fine (PM₂), coarse (PM_{10-2}) and total PM_{10} aerosols sampled a kerbside (street canyon) area in downtown Dar es Salaam. The study aimed to know the pollution levels due to metals and/or toxic elements. Principle component analysis was applied in the data set to investigate the common variability of the measured elements and to identify the emission sources.

Methodology

Sampling Protocol

The sampling campaign took place during the 2005 dry season, from 16 August and 16 September. The site is located at 6°48′51.3" S, 39°17′15.4" E, altitude 4 m a.s.l., and is at about 1 km to the Indian Ocean. A total of 51 parallel collections were made with a Gent PM₁₀ stacked filter unit (SFU) sampler, $PM_{10}(NN)$ (Hopke et al., 1997). The sampler operated at a flow rate of 17 L/min. At this flow rate the coarse filter of the SFU sampler collects the particles with aerodynamic diameter (AD) between 2 and 10 µm, while the fine filter collects the size fraction $< 2 \mu m$ AD. The diameter of the filters was 47 mm for both samplers. The samplers were placed on the second-floor balcony about 10 m above the ground level of the Young Women's Christian Association of Tanzania (YWCA) building. The samplings were conducted separately for daytime and nighttime. The 12-hour samplings started at (Local time, UTC + 3) for the daytime samples and the exchange of filters was done at 7.00 pm, for nighttime samples. Besides the actual samplings also 7 field blank

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samplings (of 30 s) were performed. The filters of the actual samples and field blanks were placed into polycarbonate Petrislide dishes and kept frozen at -20 °C during storage and transported cool to the Institute for Nuclear Sciences, Ghent University (Belgium), for analysis.

Aerosol Analyses

For Particulate mass measurement, the field blanks and actual samples were weighed before and after sampling with a Mettler MT5 microbalance (sensitivity 1 µg). Before weighing, the filters were conditioned at a relative humidity of 50% and a temperature of 20 °C for 24 hours and the weighings were done at these conditions. One quarter of each Nuclepore polycarbonate filter was used to determine black carbon (BC) by a light reflectance technique and 25 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Ba, and Pb) by particle-induced X-ray emission (PIXE) spectrometry, (Maenhaut and Cafmeyer, 1998). However, it should be noted that the PIXE data for the lightest elements (Na and Mg) are not very reliable because of severe Xray attention. The reported atmospheric concentrations for the PM₁₀ mass and elements were corrected for field blanks. In order to obtain PM_{10} data, the coarse (PM_{10-2}) and fine (PM₂) filters data were added.

Results and Discussions PM mass concentration

The median concentrations of the PM mass and elements measured in the 2005 dry season campaign in Dar es Salaam are given in Table 1 for the fine (PM_2) , coarse (PM_{10-2}) , and PM₁₀ size fractions; PM₂/PM₁₀ ratio and for PM₁₀ also concentration ranges are given. The median concentration of the PM₁₀ mass was 58 μ g/m³. In Tanzania there is no ambient PM₁₀ standard; however, the results from this study can be compared to the EU limit values. The average PM₁₀ mass concentration in Dar es Salaam is similar or higher than the daily EU PM₁₀ limit value of 50 μ g/m³ in ambient air. Actually, at this kerbside site, the daily PM₁₀ concentration exceeded the daily EU standard 38 times during the campaign. When compared with PM₁₀ data for kerbside sites in Europe, as reported in the European aerosol phenomenology study (Van Dingenen et al., 2004), the results for PM_{10} mass in Dar es Salaam are slightly higher than at the Europe sites. The ratio of PM_2 to PM_{10} averaged 0.70.

These results indicated that the PM_2 concentrations contribute the majority of the PM_{10} concentration at this site and suggests that there is higher impact of traffic at the kerbside.

Major, Minor, and Trace Elements Concentrations

The concentrations of elements for the collected air samples in Dar es Salaam are also presented in Table 1. As is commonly observed in elemental analyses of PM samples, the concentrations of the heavy metals are lower than those for the elements of crustal origin (Al, Si, Ca). Nevertheless, some typical anthropogenic metals, such as Zn and Pb, exhibited much higher median PM₁₀ levels, suggesting strong local sources for these elements in Dar es Salaam. The results also showed very strong day and night differences (hereafter D and N) for the crustal elements (Al, Si, Ca, Ti and Fe). For example, the average ratio of the PM10 level during the day to that during the subsequent night (D/N concentration ratio) was 3.3 ± 1.0 (N = 31) for Al, Si, and Fe. In contrast, the D/N ratio for the sea-salt elements, Na and Cl was 0.7 ± 0.4 and 1.1 ± 0.4 , respectively; for Zn it was $0.52 \pm$ 0.55 and for Pb 0.78 \pm 0.70. This indicates that the day/night differences for the crustal elements are not due to differences in boundary layer height between day and night, as the latter should also have affected the D/N ratio for the sea-salt elements and heavy metals. The day/night difference for the crustal elements can be attributed to resuspended road dust by vehicular traffic, which is more intense during the day. It can be seen from Table 2 that in the coarse size fraction there was a good correlation between the crustal elements and, for example, BC and Cu for which traffic can be expected to be an important source.

Fine to PM₁₀ ratios

Average fine to PM_{10} concentration ratios were calculated for the PM mass, BC, water-soluble ions and various elements; the results are shown in Figure 1. The average percentages of the PM_{10} mass in the fine (PM_2) size fraction during the campaign was 27 ± 8 %, indicating that most of the PM_{10} mass was in the coarse size fraction. Figure 1 also shows the average percentage (and associated standard deviation) of the PM_{10} aerosol in the fine size fraction for the BC, ammonium, nitrate, nss-sulphate, and several elements during the campaign. As expected, the crustal element (Al, Si, Ca, Ti, Mn, and Fe) and seasalt elements (Na, Mg, Cl, Sr) were even more associated with the coarse size fraction (for the crustal elements typically about 90%) than was the case for the PM mass; also P and Cu were mainly in the coarse size fraction. In contrast, BC, non-sea-salt (nss) S, K, V, and Pb were mostly associated with the fine size fraction, suggesting that these species and elements originated mainly from anthropogenic hightemperature sources and/or gas-to-particle conversion. Nickel on the other had about equal percentages in the two size fractions. Potassium (a well-known indicator for biomass burning) was mostly associated with the fine particles (in contrast to Ca).

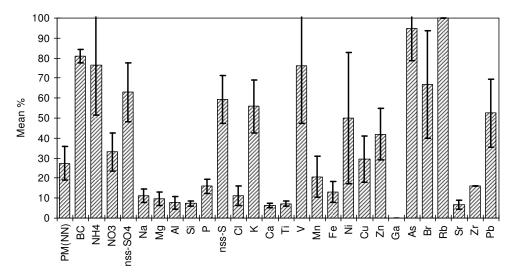


Figure 1. Mean percentage and associated standard deviation of the PM10 aerosol in the fine (PM2) size fraction for the PM mass, BC, water-soluble ions and various elements at Dar es Salaam.

Crustal Enrichment factors

Median crustal enrichment factors (EFs) for the various elements in the coarse and fine size fractions were calculated relative to the average crustal rock composition of Mason and Moore (1982), with Fe as reference element:

$$EF = (X_{PM}/Fe_{PM})/(X_{crust}/Fe_{crust})$$

where X_{PM} and Fe_{PM} are the concentrations of the measured element and Fe in the aerosol, while X_{crust} and Fe_{crust} are the concentrations of the measured element and Fe in average crustal rock composition of Mason and Moore (1982). For elements with an EF close to unity, soil dust (or other crustal matter, such as road dust) is the predominant source. If the EF is over 5, the element would have a significant contribution from non-crustal sources.

The average EFs for BC the ratio to Fe (instead of the EF) and the various elements in

the 2005 dry season campaign are shown in Figure 2 and 3 for the fine and coarse size fractions, respectively. Most elements in the coarse size fraction have EFs very close to one, suggesting that they are mainly thus attributable to soil dust dispersal and road dust re-suspension. In contrast, Ca exhibits high EFs in both size fractions. This is attributable to enrichment of the local soil (and road dust) this site with limestone soil. at The enrichments for Na and Cl are due to the fact that these elements have sea-salt as their major source. Several elements, including S, K, V, Ni, Cu, Zn, As, Br, Rb, and Pb have high to very high EFs in the fine size fraction, which suggests that they were mainly of anthropogenic origin and that their sources were within the city, such as traffic.

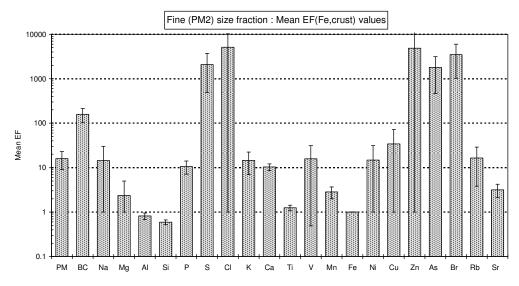


Figure 2. Mean crustal enrichment factors, calculated relative to the average rock composition of Mason and Moore (1982) for the **fine** size fraction aerosols in Dar es Salaam. The error bars denote 1 standard deviation.

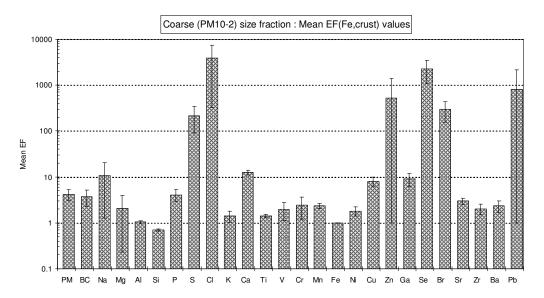


Figure 3. Mean crustal enrichment factors, calculated relative to the average rock composition of Mason and Moore (1982) for the **coarse** size fraction aerosols in Dar es Salaam. The error bars denote 1 standard deviation.

Sources of certain Elements and Aerosol Components

From the concentrations of the crustal elements, the concentration of the crustal (matter) aerosol component was calculated and it accounted for about one third of the PM_{10} mass. A large fraction of the crustal component is undoubtedly due to resuspended road dust, particularly during the daytime, when the concentrations of the crustal elements and traffic intensity were much greater than during the night. During this 2005 dry season campaign, the concentration of the

crustal component in PM₁₀ was, on average, 35 μ g/m³ during the daytime and only 10 μ g/m³ during the night; for comparison, the sea-salt concentration (calculated from Na and Cl), was very similar during day and night (on average, around 9 - 10 μ g/m³). It is therefore reasonable to assume that the day/night concentration difference of 25 μ g/m³ for the crustal matter component is essentially due to road dust. The difference between the average daytime and nighttime PM₁₀ mass concentrations in the same campaign was 49 μ g/m³, of which 25 μ g/m³ came thus from crustal matter. The remaining 24 μ g/m³ is likely also partly related

to traffic and may to a large extent consist of carbonaceous matter.

To elucidate the sources of the various elements at Dar es Salaam, crustal enrichment factors (see Figures 3 and 4) and pairwise correlation coefficients were calculated and various scatter plots were constructed. Potassium was substantially enriched in the fine size fraction, on average, around 10, which is similar as observed at African sites that were heavily impacted by biomass burning and where the pyrogenic aerosols accounted for a large fraction of the PM mass (Maenhaut et al., 1996). This suggests that biomass burning (in this case for cooking) was also a main contributor to the PM mass at the Dar es Salaam site. Fine V and fine Ni were highly correlated with each other $(r^2 = 0.76)$ and their concentration ratio was around 2, which is consistent with residual oil burning as a source for these elements. BC and coarse Cu were highly correlated with the crustal elements, suggesting that they were related to traffic sources. For BC. automotive exhaust emissions, in particular from diesel vehicles, are undoubtedly important. That also Cu comes from traffic is not unexpected, as abrasion of brake linings wear is a well-known source for this element. Also coarse Zn and fine Pb are in cities often related to traffic, with tire wear and leaded gasoline as important sources, respectively (Ruellan and Cachier, 2001). However, neither Zn nor Pb was strongly correlated with the crustal elements (or with any other element) at Dar es Salaam; besides, the ratio of non-sea-salt Br to Pb was much lower than in particulate emissions from leaded gasoline. It is therefore thought that Zn and Pb originated from industrial emissions and possibly also re-suspended contaminated road dust, and that different industries were responsible for the two elements. For Pb this could be a battery industry and for Zn several metal processing industries, which are all located within 10 km from the sampling site.

Correlation between Various Aerosol Components

The correlation matrix for the PM mass, BC, various elements, in the fine (PM2) and the coarse particle (PM10-2) size fractions were calculated. Most elements exhibit strong correlations in the coarse size fraction and somewhat weaker ones in the fine size fraction. This suggests that many elements originate predominantly from the same source.

In the fine particles, a higher correlation was observed for BC and most of the crustal elements, such as Al, Si, Ca, Ti, and Fe (r \geq 0.73), indicating that they may be associated with traffic and resuspended road dust. Also for the coarse size fraction significant correlations existed between BC and the pollution elements Ni and Cu ($r \ge 93$) and with crustal sources elements (r \geq 96). The correlation between Ni and Cu in the coarse size fraction was r = 0.90; and both of them were also strong correlated with typical crustal elements, BC, K, and the PM mass ($r \ge 0.88$). This all could be related to the resuspension from road dusts caused by moving vehicles and exhaust gases.

Source Identification by Factor Analysis

In this work, Principal Component Analysis (PCA) was applied to the fine and coarse size fraction data sets. Briefly, PCA makes use of an eigenanalysis of the correlation matrix of the data set, after which a limited number of principal components (PCs) and their associated eigenvectors are retained and a rotation of the matrix with retained eigenvectors is carried out (Maenhaut and Cafmeyer, 1987). The most commonly used rotation is the VARIMAX rotation of Kaiser (1958) and this rotation was also used here. The identification and "naming" of the VARIMAX rotated PCs in terms of aerosol sources (or source types) is based on the loadings for the various variables within each PC.

Principal Component Analysis on the Fine Size Fraction

Twenty variables, i.e., PM, BC, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Br, Sr and Pb, were included for the PCA on the fine size fractions data sets. The VARIMAX rotated PCA resulted in five PCs, for which the loadings are given in Table 2. The five components explained 86.5% of the variance in the data set. The first component (Comp 1) is very highly loaded with the crustal elements Al, Si, Ca, Ti, Mn and Fe and is clearly a crustal component; it represent road dust. The second component has very high loadings for Na and Mg and high loadings for Br. It represents undoubtedly sea salt. Comp 3 is highly loaded with the potential pollution elements Zn and As and represent likely different pollution sources within Dar es Salaam. These pollution sources include several metal processing industries such as

metallurgical factories, alloy industries, galvanizing activities in industries, and galvanized parts of heating appliances could be the main sources for Zn and As. Comp 4 is very highly loaded with K; besides, S is highly correlated with this component. The high loading for K indicates that this component contains biomass burning aerosol. Also the S may in part be associated to biomass burning. The fifth component (Comp 5) is unique components, since only one species is highly loaded on it. BC is highly loaded on it, as are the D_N variable. Comp 5 is therefore also associated with the daytime and with emissions from traffic.

Principal Component Analysis on the Coarse Size Fraction

For the coarse size fraction data set from the 2005 dry season campaign at Dar es Salaam 23 variables were included in the PCA, i.e., the same variables as for the fine data set, and in addition Ni, Ga and Ba. Five PCs were retained, which explained 90.7% of the total variance of the data set. The VARIMAX rotated PC loadings are given in Table 3. Several variables are very highly or highly loaded on the first component, which seems to be the coarse fraction counterpart of Comp 1 in the fine size fraction. BC is also highly loaded on it, as are the Cr and Ni. Comp 1 is therefore represents a mixture of road dust and traffic exhaust. That Cu is also very loaded on this component may be due to the contribution from brake lining wear in automobiles. It is well-known that this process is an important source of coarse Cu at kerbsides (e.g., Salma

and Maenhaut, 2006; and references therein). Comp 2 is very highly loaded with Na, Mg, and Cl; represents sea salt, similarly as Comp 2 in the fine size fraction. The third coarse component is unique highly loaded with Pb and to some extent also with Ba. It clearly stands for pollution component and the battery industry in Dar es Salaam could be the main source of Pb. The final component (Comp 4) is loaded with Zn and it is undoubtedly the counterparts of the Comp 5 in the fine size fraction.

Conclusion

The study of the atmosphere particles from Dar es Salaam revealed the presence of major, minor and trace elements, leading to the characterisation of different sources: the crustal, sea spray, biomass burning, traffic, and pollution sources. The PM mass concentrations obtained in Dar es Salaam were similar or higher than the daily EU PM₁₀ limit value of $50 \ \mu g/m^3$ in ambient air. The presence of trace elements Ni, Cu and Zn in the fine fraction of the particulate, along with S, V, Cr and Br, was verified through the enrichment factor, showing values characteristic of anthropogenic contribution. Principal component analysis with VARIMAX rotation was applied to the separate fine and coarse size fraction. Five components were identified for the fine size fraction and explained 86.5% of the variance in the data set. While in the coarse size fraction four components were identified and explained 90.8% of the total variance of the data set.

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Table 1. Median concentrations (ng/m^3) in the fine (<2 μ m AD), coarse (10–2 μ m AD) and PM10 size fractions (and concentration ranges for PM10) at Azikiwe Street site, Dar es Salaam during the 2005 dry season campaign (based on 64 samplings).

	Fine	Coarse	PM10	PM10 conce	ntration range	
Species	Median	Median	Median	Min.	Max.	PM2/PM10 Ratio
PM	16930	40040	58000	23000	130000	0.29
Na(IC)	300	2602	3000	1170	6900	0.10
Mg(IC)	35.5	370	420	220	910	0.08
Al	59.09	549.5	620	170	1920	0.10
Si	163.6	1806	1990	550	6300	0.08
Р	15.38	46.1	47	DL	160	0.33
S	466.6	586	1080	580	2800	0.43
Cl(IC)	465	4260	3900	1200	9200	0.12
K(IC)	379	299	650	290	1450	0.58
Ca(IC)	423	5920	4600	1140	16900	0.09
Ti	5.615	58.74	65	16	200	0.09
V	1.232	1.677	0.87	DL	10	1.42
Cr	5.17	3.51	1.7	DL	9.0	3.04
Mn	3.478	20.09	24	4.7	79	0.14
Fe	53.94	449	500	141	1700	0.11
Ni	0.8067	1.7285	1.5	DL	7.3	0.54
Cu	1.718	4.163	7.5	2.0	21	0.23
Zn	133.6	134.2	270	22	3200	0.49
Ga	DL	1.829	1.3	DL	4.7	DL
As	3.4145	3.655	1.6	DL	31	2.13
Se	2.0585	2.007	1.6	DL	11	1.29
Br	7.962	8.102	12	3.3	36	0.66
Rb	1.759	5.266	DL	DL	5.3	DL
Sr	1.687	10.82	11	1.9	35	0.15
Ba	DL	17.55	DL	DL	30	DL
Pb	62.14	46.62	110	1.9	1090	0.56

DL - detection limit; IC - ion chromatography.

Variable	Comp 1	Comp 2	Comp 3	Comp 4	Comp 5	Communalities	
	Crustal	Sea spray	Pollution	biomass burning	traffic		
PM	0.34	0.35	-0.17	0.64	0.48	0.91	
BC	0.21	0.10	-0.17	0.20	0.74	0.67	
Na	-0.15	0.89	-0.16	0.33	0.11	0.95	
Mg	-0.10	0.88	-0.26	0.24	0.10	0.92	
Al	0.98	-0.04	-0.09	0.05	-0.02	0.98	
Si	0.98	-0.06	-0.06	0.08	0.02	0.97	
Р	0.79	-0.10	-0.35	0.13	-0.36	0.90	
S	0.15	0.23	-0.26	0.86	0.24	0.95	
Cl	-0.10	0.67	0.55	-0.45	0.06	0.98	
К	0.14	0.24	-0.24	0.87	0.27	0.95	
Ca	0.93	-0.07	0.01	-0.08	0.13	0.89	
Ti	0.96	-0.05	0.02	0.05	-0.01	0.93	
Mn	0.91	0.03	0.21	0.03	0.08	0.89	
Fe	0.96	-0.02	-0.02	0.11	0.11	0.95	
Cu	-0.07	0.12	0.34	0.61	-0.11	0.52	
Zn	0.20	0.04	0.90	-0.21	-0.05	0.90	
As	-0.27	-0.08	0.86	0.05	0.02	0.83	
Br	0.02	0.86	0.24	0.29	-0.21	0.93	
Sr	0.57	0.16	-0.48	0.27	-0.17	0.68	
Pb	0.16	-0.11	0.64	0.02	-0.51	0.70	
Variance	6.9	3.2	3.2	2.9	1.9		
% Variance	32.7	15.5	15.2	14.2	8.9		

Table 2. VARIMAX rotated PC matrix for the fine samples from the 2005 dry season campaign at Dar es Salaam. High (>0.7) and very high (>0.9) loadings are indicated in bold.

Variable	Comp 1	Comp 2	Comp 3	Comp 4	Communalities	
	Crustal and	Sea	Pb	Zn	1	
	traffic	spray	10			
PM	0.93	0.02	0.27	-0.11	0.96	
BC	0.90	-0.08	0.09	-0.10	0.84	
Na	-0.18	0.96	-0.03	-0.06	0.97	
Mg	-0.10	0.98	0.04	-0.13	0.99	
Al	0.94	-0.28	0.11	-0.16	0.99	
Si	0.92	-0.29	0.14	-0.19	0.98	
Р	0.89	0.00	0.03	-0.28	0.88	
S	0.80	0.48	0.25	0.05	0.93	
Cl	-0.02	0.95	-0.18	0.08	0.94	
K	0.55	-0.04	0.65	-0.22	0.78	
Ca	0.96	-0.21	0.04	-0.09	0.97	
Ti	0.87	-0.34	0.25	-0.25	0.99	
Cr	0.74	0.12	-0.45	-0.03	0.76	
Mn	0.88	-0.27	0.24	-0.23	0.95	
Fe	0.93	-0.24	0.19	-0.11	0.97	
Ni	0.84	0.45	0.03	-0.24	0.96	
Cu	0.86	0.12	0.25	0.16	0.84	
Zn	-0.02	-0.09	0.11	0.92	0.86	
Ga	0.98	-0.01	0.08	0.09	0.98	
Br	-0.46	0.03	-0.04	0.73	0.75	
Sr	0.96	0.09	0.05	-0.20	0.97	
Ba	0.40	0.08	0.79	0.00	0.79	
Pb	-0.10	-0.28	0.74	0.44	0.83	
Variance	12.70	3.80	2.20	2.10		
%	55.40	16.50	9.80	9.00		
Variance						

Table 3. VARIMAX rotated PC matrix for the coarse samples from the 2005 dry season campaign at Dar es Salaam. High (>0.7) and very high (>0.9) loadings are indicated in bold