Full paper

Aerosol Characterization for Minna, Nigeria By EDXRF Technique And Source Attribution By PCFA And CMB Multivariate Statistical Techniques

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ABSTRACT

The ambient aerosols sampled gravimetrically in Minna during a dry season were analyzed using an ED-XRF system. Principal Component Factor Analysis (PCFA) and Chemical Mass Balance (CMB) statistical techniques were used to infer source of the aerosols and their contributions.

The average aerosols concentrations at the city centre and residential areas were respectively 288 - 483 μg/m^3 (pre-harmattan) and 100 – 111 μg/m^3 (pre-harmattan, but 362 -1200 μg/m^3 during harmattan). Al, Si, S, K, P, Ca, Ti, Fe, Cu, Zn, V, As, Sr, Pb, Cr, Mn, Co, and Br were detected but only Zn and Pb were highly/moderately enriched. Computed elemental ratios of Pb/Zn and Pb/Br from the samples indicated high vehicular transportation contributions in the commercial areas. However K/Fe ratios showed high wood/biomass burning contributions during the daytime. The inter-elemental correlation showed significant ($r^2 > 0.5$) associations between (Al, Si, Fe, Mn, and S), (K, Ti, and Zn), (Pb, P, and Cu), and (Br, Ca, Cu and P), indicating a complex source interactions. Loadings from the PCFA were: Factor 1 suggesting soil source; Factor 2 suggesting automobile source; Factor 3 suggesting harmattan and entrained soil from vehicular dust remobilization. A spread of wood burning influence over Factors 2 and 3 was noted. The CMB analysis provided mass contributions from Factors 1 to 3 to be 52, 28 and 21 % respectively. The study showed insignificant contribution of local industries to the observed aerosol composition in the city. Salient air quality management options for the city airshed were however discussed.

1. INTRODUCTION

Much of the published aerosol studies in Nigeria have focused on the industrial/urban cities such as Lagos (Ogunsola *et al*, 1994 a; Baumbach *et al*, 1995; Oluwole, *et al*, 1994; and Ogunsola *et al*, 1994 b), Ibadan (Oluwande, 1977 and Oluwande, 1979), and Kano (Beavington and Cawse, 1978). Relatively very little ambient aerosol studies have been done at non-industrial 'middle belt' urban areas of the country, an ecological zone to which the nation's new capital (Abuja) belongs. A second reason is to investigate to what degree the "marker elements" for the local industry in Minna can be fingerprinted in the ambient aerosol. These reasons, among others, informed the selection of Minna for this study. In an earlier review, particulate matter has been identified as the aerosol constituent of major concern in the Nigerian ambient environment (Akeredolu, 1989). The multi-elemental capability of the ED-XRF technique, and its suitability for urban aerosol studies has been demonstrated in earlier studies (Ogunsola *et al*, 1994 a) while the strong contribution by harmattan dust to air quality in Nigeria has been documented by Asubiojo *et al* (1994).

Minna city (latitude 9^{o}37N, longitude 6^{o}33E) has metamorphosed from the time when it was first settled by rail construction worker in 1905 till it became, in 1976, the state capital of the newly created Niger State. The city is widely dispersed along the main spine road (F 126) from Chanchaga in the South to Bosso in the North (Figures. 1 and 2). The main Kano – Lagos rail line runs east west, almost bisecting the city. Minna city covers some 900 hectares comprising low, medium and high density residential areas which respectively 14.3, 20.6 and 19.7 % of the total land use whilst institutional land use accounts for 45% (Niger State, 1980). The city now boasts of a Federal University of Technology and an international airport. Road dualization projects have also been carried out there. These facts point to the rapid urbanization occurring there. It is located on a geological base of undifferentiated basement complex mainly gneiss and magnetite. The city enjoys a climate typical of the Middle belt zone of Nigeria.

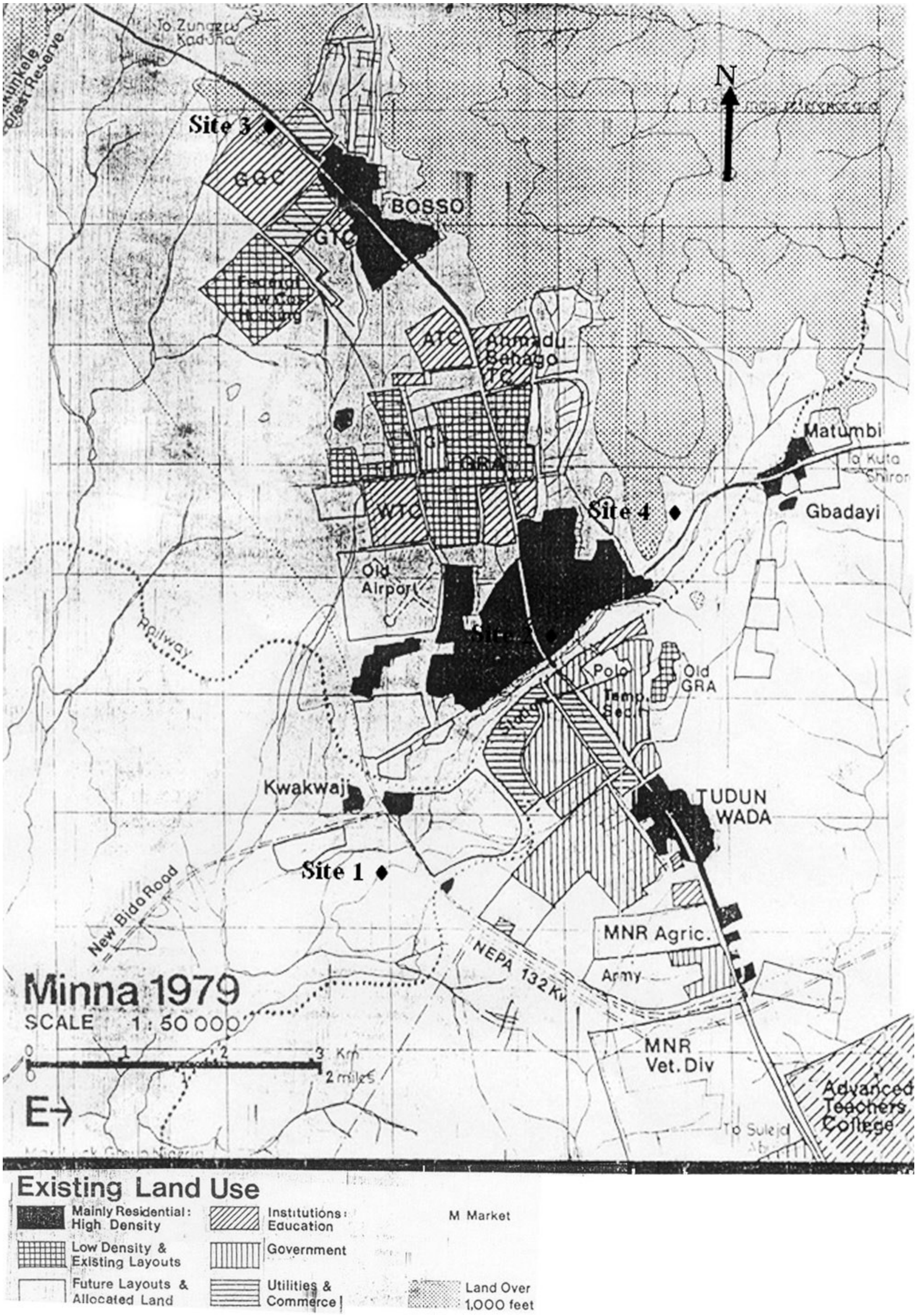

Figure 1: Map of Minna Showing the Land Use and Air Sampling Sites

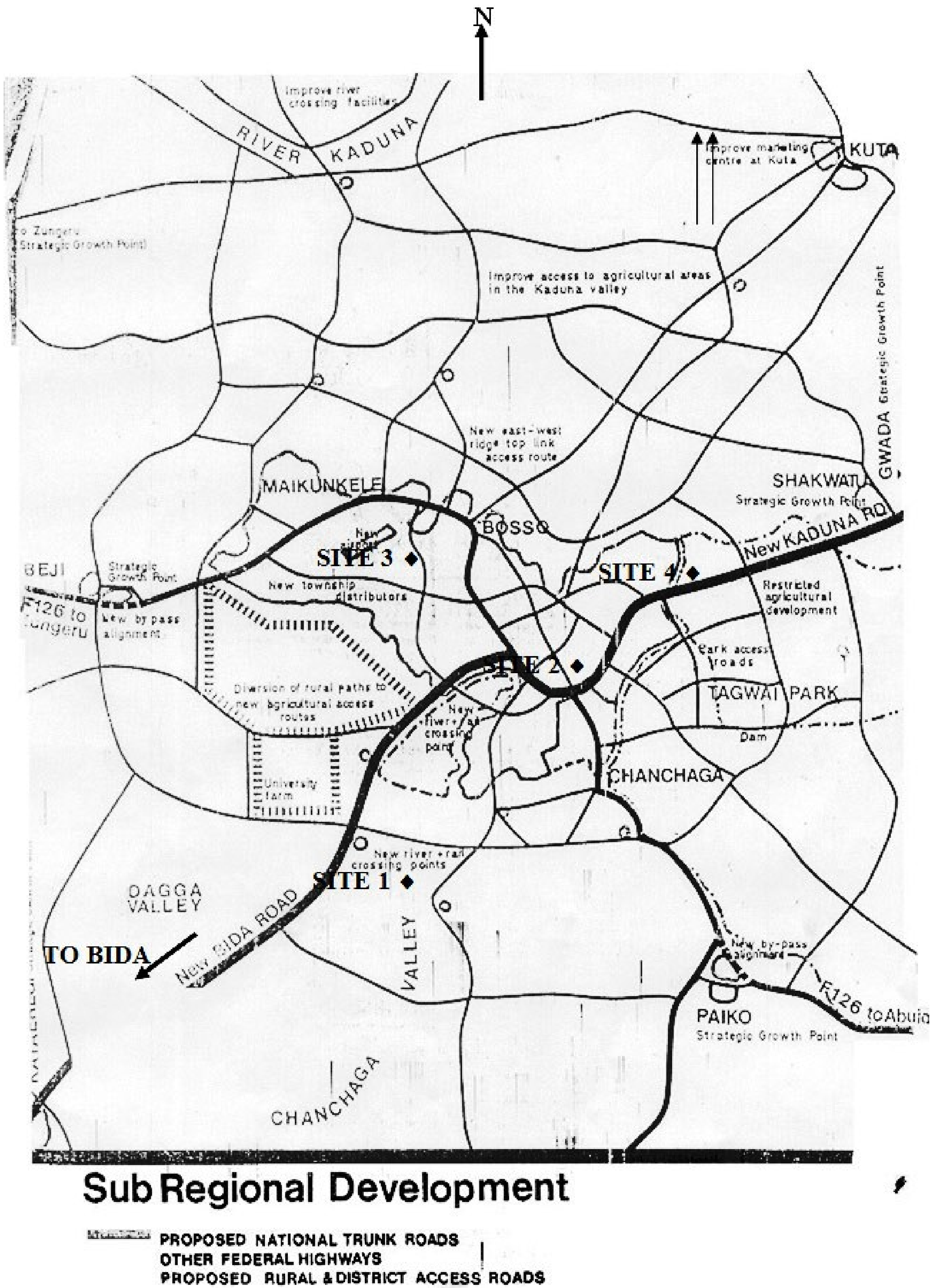

Figure 2: Map of Minna and Sub Regional Development with Air Sampling Sites

Its rainy season lasts 190 to 200 days with a mean total annual rainfall of 1334 mm and its highest rains of 300 mm occurring in September. The mean monthly temperature is highest in March at 30.5°C and lowest in August at 22.3°C (Niger State, 1986). The mean monthly wind speed (expressed in terms of "wind run") reported for Minna ranges between 9 and 115 km (Niger State, 1990).

Air pollution sources identified in Minna include transportation, solid waste burning, bush burning, harmattan and construction and the local brass foundry. This paper reports the concentration and elemental compositions of total suspended particulate matter collected in the city during a dry season air sampling campaign that took place in 1994.

1.1. Local Industrial Activity in Minna City and Environs

Minna, which was a small-scale gold mining town, is now being transformed into an industrial centre (Niger State, 1986). At Pago, 17km SE of Minna is located a burnt-bricks factory with installed capacity for producing 300million bricks/year. Other major modern industries in the area include Wire Products industry, a Fruit Juice Factory, a Marble Factory, and a Liquefied Petroleum Gas Limited. Small scale ventures include sawmills, marble quarry, furniture making (Niger State, 1986), and "blacksmithing" (Niger State, 1990). Bida, a town located southwest (i.e. upwind) of Minna (Figure 2), contains brass foundries. There was interest in ascertaining whether the local brass foundries and brass-working industry leave any noticeable footprints in the aerosol that was sampled at Minna the elemental ratios were therefore examined to find out if the ratios of marker elements found in brass correspond with those occurring in the aerosol.

2. MATERIALS AND METHODS

The gravimetric air sampling was conducted using a portable low volume air sampler at two classes of locations within the city: the Central Business District (commercial areas) and the Residential areas (low density residential areas). The sampler is battery operated at a flow rate of 10 litres per minute and runs between 6-8 hours. Particles were collected on pre-weighed Whatman 41 cellulose filters and final weights determined after sampling. The mass difference divided by volume of air sampled resulted in the TSP concentration. The filters were subsequently analyzed using an ED-XRF multi-elemental analysis system.

2.1. Analytical Technique

The aerosol samples collected on filters were analysed in 1994 by means of an Energy Dispersive X-ray Fluorescence (EDXRF) system Link Analytical XR 300 model coupled with computer based MCA which controls the sample holder and data acquisition. The X-ray beam is produced from a tube with Rh anode. Si (Li) semiconductor detector detects the characteristic X-ray from sample at 90° relative to the incident rays. Calibration of the system was done using single element standards provided by the manufacturer. Samples were mounted on disposable XRF sample holders and measured for 600s with an accelerating voltage of 20 kV and a current of 0.10 mA. The analysis was carried out at these settings because the elements of interest have their characteristic X-rays within the energy range selected. Fundamental parameter method was used for elemental concentration determination. The elemental Enrichment Factors (E.F) were calculated from the elemental compositions so determined using Eqn. (1) with Ti as the normalizing element:

$$EF = \frac{\dfrac{(weight\%\ of\ element\ i\ in\ sample)}{(weight\%\ of\ \mathrm{Ti}\ in\ the\ sample)}}{\left(\dfrac{weight\%\ of\ element\ i\ in\ soil}{(weight\%\ of\ \mathrm{Ti}\ in\ the\ soil)}\right)} \quad (1)$$

These compositions were fed into the Factor Analysis (both Principal Component and Varimax rotation methods) and the Chemical Mass Balance (CMB) receptor models for source identification and apportionment purposes. Appendix I give the theoretical basis of the Principal Component Factor Analysis tool employed in this work.

3. RESULTS AND DISCUSSION

The gross concentrations of the total particulate matter measured are as shown in Table 1. The average TSP concentrations measured at the city centre, commercial district and residential areas were respectively 288 – 483 μg/m^3 (pre-harmattan) and 100 – 111 μg/mm^3 (pre-harmattan but 362 –1200 μg/m^3 during harmattan episodes). These were all higher than the 250μg/m^3 daily average of national ambient air quality standards of the Federal Ministry of Environment (FEPA, 1991) except the concentrations at the commercial district before the harmattan.

Table 1: Ambient Air Particulate Concentration in Minna Aerosols Sampled

Sample No.	TSP concentration (μg/m^3)	Remarks	
	City Centre Samples		
1	458	Non-harmattan samples; Within	
2	288	20m of busy road artery.	
3	483		
	Residential Area Samples		
4	100	Non-harmattan samples	
5	362		Daytime
6	625		Nighttime
7	700	Harmattan	Daytime
8	1200	samples	Nighttime
9	650		Daytime

Typical elemental compositions found in the aerosols and the corresponding enrichment factors are summarized in Table 2. The inter-elemental correlation (Table 3) showed that significant ($r^2 > 0.5$) associations exist between (Al, Si, Fe, Mn and S), (K, Ti and Zn), (Pb, P, and Cu), and (Br, Ca, Cu and P), for instance. These associations, if they were real were indicative of complex source interactions in the aerosol thus the need for multivariate statistical techniques (PCFA and CMB) to infer the sources of the aerosols and their mass contributions to the sampled aerosol.

From the Factor Analysis results (Table 4), three principal component factors were found significant with the following loadings: Factor 1 is suggestive of soil with high loadings of Al, Si, Fe, Mn and S. Factor 2 is suggestive of automobile sources with high loadings of Pb, P and Cu, and moderate loadings of Br. and S. Factor 3 is suggestive of harmattan and entrained soil due to vehicular dust remobilization with high loadings of Ca, and Br and moderate loadings of S and Zn. There appeared to be a spread of wood burning influence over Factors 2 and 3. Mass contributions by Factors 1 to 3 found from the CMB analysis were respectively 52, 28 and 21 %.

The elemental ratios were examined in order to find out if the ratios of marker elements found in brass correspond with those occurring in the aerosol. The ratio of K/Fe ranged between 3.26 and 10.7 which implied that there appeared to be a contribution from biomass burning to the aerosol while Cu/Zn ratio ranged between 0.0029 and 0.0042. By comparison, Leidheiser (1971) reported Cu/Zn ratio in nine different commercial brass alloys to range between 1.5 and 5.62. Thus, fumes from commercial brass production may not have strong contributions to the sampled aerosol. The Pb/Br ratio ranged from 2.68 to 2.81 which are comparable with those in samples collected near heavy traffic roads in Ile-Ife and Lagos (Table 5). The Pb/Br ratio in automobile exhaust was reported to be 2.41 - 2.56 (Harrison and Sturges, 1983 and Faiq and Allah, 1988). After making allowance for Br evaporation from the filters, the Pb/Br ratio is comparable with that in gasoline. Vanadium was found to be below the analytical detection limits in all the samples collected.

Table 2: Typical elemental composition and enrichment factors of Minna aerosols

ELEMENT	City Centre (Sample #3)		Residential Area (Non-Harmattan sample #4)		Residential Area (Harmattan Episode Samples)			
					Sample #6		Sample #9	
	Concn., (%)	E.F	Concn., (%)	E.F	Concn., (%)	E.F	Concn., (%)	E.F
Al	3.12	0.48	2.05	0.32	4.62	0.67	3.98	0.77
Si	23.04	0.1	23.87	0.10	27.73	0.11	29.03	0.12
P	0.55	0.18	0.44	0.15	0.52	0.13	0.49	0.12
K	4.18	0.38	4.16	0.38	3.97	0.39	3.61	0.38
Ca	4.17	1.87	2.51	1.12	3.17	1.55	3.25	1.56
Ti	2.32	1	2.33	1.00	2.18	1.00	2.03	1.00
Fe	0.62	0.13	0.20	0.04	1.24	0.28	1.03	0.30
Cu	62 ppm	0.13	59 ppm	0.10	78 ppm	0.00	58 ppm	0.61
Zn	2.07	35.56	1.99	34.06	1.92	37.08	1.87	37.21
As	127 ppm	0	0	0.00	120 ppm	0.00	117 ppm	0,00
Sr	233 ppm	0.27	258 ppm	0.29	203 ppm	0.27	229 ppm	0.29
Pb	247 ppm	4.59	218 ppm	4.03	224 ppm	0.00	214 ppm	0.00
Mn	441 ppm	0.23	235 ppm	0.12	618 ppm	0.35	448 ppm	0.31
S	0.36	2.17	0.1527	0.91	0.4734	2.92	0.41	2.57
Co	79 ppm	0	67 ppm	0.00	85 ppm	0.00	81 ppm	0.00
Br	88 ppm	0	0	0.00	83 ppm	0.00	80 ppm	0.00
Cl	ND	0	ND	0.00	0.15	2.75	0.15	3.01

Table 3: the inter elemental correlation for the aerosols sampled at minna

	Al	Si	P	K	Ca	Ti	Fe	Cu	Zn	Pb	Mn	S	Br
Al	**1.00000**												
Si	0.80540	**1.00000**											
P	0.34747	0.07845	**1.00000**										
K	-0.6193	-0.73618	0.19536	**1.00000**									
Ca	-0.10107	-0.45845	0.308884	0.27293	**1.00000**								
Ti	-0.62122	-0.69427	0.24000	0.98878	0.23968	**1.00000**							
Fe	0.97627	0.83585	0.17792	-0.70764	-0.10912	-0.72112	**1.00000**						
Cu	0.22992	0.13308	0.89628	0.30901	0.04150	0.36800	0.06331	**1.00000**					
Zn	-0.59191	-0.67954	0.13843	0.92766	0.44383	0.92992	-0.65367	0.16978	**1.00000**				
Pb	0.22910	0.28157	0.65687	0.11958	-0.21803	0.20079	0.13503	0.67283	0.08680	**1.00000**			
Mn	0.77523	0.58905	-0.05388	-0.49725	0.10260	-0.53952	0.85827	-0.13626	-0.41770	-0.07089	**1.00000**		
S	0.85343	0.58367	0.52802	-0.27738	0.24724	-0.29487	0.80197	0.34966	-0.17092	0.27818	0.64617	**1.00000**	
Br	0.16522	0.05078	0.65884	0.04235	0.66315	0.02246	0.12766	0.49928	0.06636	0.21078	0.09122	0.43949	**1.00000**

3.1. Comparison of Results of Day-time sampling with those of Night-time sampling

The results obtained during daytime sampling were compared with nighttime sampling as follows: The elemental enrichment factors were used rather than elemental concentrations; the hypothesis tested was that daytime and night time results were identical while paired values EF for the 13 elements were analyzed. Similarly, the difference d_i between the i^{th} pair of observations was calculated using Equation (2).

$$d_i = \left(\frac{EF_{day,i} - EF_{night,i}}{EF_{day,i}} \right) \quad (2)$$

In order to decipher the differences between the emission sources predominating during the daytime/night sampling, the K/Fe ratios (markers for wood burning) were compared (Table 5) for these and other elemental ratios. Daytime values of that ratio ranged between 3.20 and 2.06 whilst overnight ratios ranged between 2.97 to 4.1. There was a statistically significant difference between those ratios. It was inferred that more refuse/wood combustion take place during daytime is reflected in the K/Fe ratios.

3.2. Investigation of Transportation Contributions (Using Pb/Br ratio):

The Pb/Br ratios of 2.68 to 2.81 were found comparable with Pb/Br of 2.56 and 2.24 respectively reported for US and Nigerian vehicle exhaust (Ogunsola *et al*, 1993). It was therefore inferred that vehicle exhaust emission from transportation was a very likely source of aerosols at Minna.

Table 4: Principal Component Factor Analysis results for Minna Aerosol samples (Rotation Method: Varimax)

Orthogonal Transformation Matrix

	1	2	3
1	0.99561	0.09280	-0.01247
2	-0.07172	0.84138	0.53566
3	0.06020	-0.53241	0.84434

Rotated Method: Varimax Rotated Factor Pattern

	FACTOR 1	FACTOR 2	FACTOR 3
Al	0.90337	0.31132	0.14982
Si	0.86962	0.25853	-0.26237
P	0.01977	0.86974	0.39793
K	-0.87362	0.23522	0.19847
Ca	-0.20461	-0.10857	0.94220
Ti	-0.88201	0.30471	0.14272
Fe	0.95946	0.15363	0.13405
Cu	-0.08361	0.92985	0.14718
Zn	-0.81681	0.13849	0.33109
Pb	0.02394	0.87685	-0.17663
Mn	0.78555	-0.10395	0.31155
S	0.64898	0.42244	0.49664
Br	0.06368	0.37407	0.74887
	Soil	Vehicular	Entrained Soil/Wood burning/Harmattan

Variance Explained by Each Factor

FACTOR 1	FACTOR 2	FACTOR 3
5.793348	3.085641	2.282090

Final Communality Estimates: Total = 11.161079

Al	**Si**	**P**	**K**	**Ca**	**Ti**	**Fe**
0.935440	0.891923	0.915186	0.857936	0.941387	0.891160	0.962127
Cu	**Zn**	**Pb**	**Mn**	**S**	**Br**	
0.893278	0.795973	0.800644	0.724957	0.846285	0.704784	

Table 5: Comparison of the Elemental Rates computed in the study with other results

Ratio	Calculated Ratio			
	Minna[1]	Bajoga[2]	Ile-Ife[3]	Remark
Pb/Br	2.68 - 2.81	-	2.12	2.24 Lagos[4]
K/Fe	3.20 - 3.50 (Harmattan) 6.12 - 20.6 (non-Harmattan)	0.14 - 0.5	0.4 - 6.5	
Pb/Zn	0.011 - 0.012	No Data		Lagos Road sides 2.21 - 5.62
Cu/Zn	0.003 - 0.004	0.4 - 1.3		Brass Alloys Cu/Zn ≈2 to 4
Ca/Si	0.11 - 0.18	0.03 - 0.26		2.61 - 5.72[5]

[1] This Study
[2] Field work by the authors (See Adejumo, 1994)
[3] Unpublished Field Work by Akeredolu (1993)
[4] Ogunsola *et al*, 1993
[5] Unpublished Field Work by Akeredolu (1995)

3.3. Investigation of Wood Burning Contributions (Using K/Fe ratio)

Potassium is commonly accepted as a marker element for aerosols of wood origin. Similarly, K/Fe ratio is commonly accepted as a good indicator of wood burning contributions. According to Dasch (1992) K/Fe > 3 indicates significant inputs from wood burning. Using this criterion, samples could be separated into two classes including: non-harmattan samples with K/Fe = 1.67 to 2.06 and harmattan episode samples with K/Fe = 3.2 to 3.5.

3.4. Investigation of Construction Industry Contributions (Using Ca/Si)

The Ca/Si ratio has been shown to portray cement (i.e. construction) contributions in previous studies (Akeredolu *et al*, 1994). This ratio was approximately 0.1 - 0.2 at Minna. By contrast, the value of the ratio in Lagos was 2.61 to 5.72 (Akeredolu, 1995). The ratio also contrasted with that found in harmattan aerosols (Asubiojo *et al*, 1994).

3.5. Investigation of Possible Local Industry Contributions

Comparison of the Cu/Zn ratio found in Minna aerosol sample (0.003 to 0.004) was made with those in commercial brass alloys published by Ledheiser (1971). It was inferred that the brass working in the area was not the main source of Cu and Zn in the aerosol.

3.6. Airshed Air Quality Management Implications of this Work

Biomass burning and open-waste burning marker elements showed very prominently in the daytime samples. Therefore, one of the options for air quality improvement is to reduce waste burning at the city's roadsides. Biodegradable constituents of the wastes can be composted, for instance. The air quality was adequate in the residential area but much poorer in the central business districts of the city, exceeding the FEPA limits in the latter. Thus, passing a bylaw in the commercial district area forbidding waste burning and enforcing solid waste collection by a specially designated solid waste management board is recommended.

Over half of the airborne dust is of crustal (soil) origin. There may be need to engage in road sweeping to reduce amount of dust available for mobilization by wind or vehicular activities. The fact that the Pb/Br ratios were identical to those found in Lagos vehicular exhaust contributions to the air quality suggested that the requirements for "road worthiness" for vehicles may need to include tail pipe emission criterion.

4. CONCLUSIONS

The TSP concentrations measured in the city-centre exceeded the FEPA limit of 250 μg/m^3 during the sampling period however at the residential areas the concentrations were below the FEPA limits except during harmattan episodes when the concentrations went as high as 4 folds of the limit. From the enrichment factors of elements and elemental ratios, it could be deduced that vehicular transport contributions were high in the commercial areas but not so high in the residential zone. Also there was a significant difference between daytime and overnight samples which suggests that the present levels of human activity perturbed the air quality significantly in that airshed. Wood and refuse mostly influenced the daytime aerosols burning.

The policy implication of this is the need to curtail open refuse burning at the street corners. The multivariate statistical analyses carried out signify that there is a complex pollution source interactions in the airshed. However, the analyses did not detect significant contribution by local industry sources in the airshed. Judging by the current air quality trends the air shed can accommodate the siting of industries without fear of significant deterioration of air quality. However, emission of dust by such industries would require to be regulated.

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APPENDIX I

Theory

In Principal Component Factor Analysis (PCFA), the raw data (X_I) are first transformed into standardized variables (Z_{ij}) in order for all variables to be treated on an equivalent basis.

i.e. $Z_{ij} = (X_{ij} - X_i)/S_i$

where X_i = mean

S_i =standard deviation of the variables

The standardized variable is represented as a product of two cofactors (i) the factor loading and (a_{ik}) which represent the correlation coefficients between the factors and the original variables and (ii) the factor scores (f_{kj}).

$$X_{ij} = \sum_{k=1}^{p} a_{ik} f_{kj}$$

Commercial PCFA algorithms determine the parameters from the parameters of the above equation. This work has used the PCFA algorithm released by SAS.