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Compositional and Air-mass Trajectory Analysis of a Heavy Dust Episode (HDE) Aerosols in Ile-Ife, Nigeria

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Authors' contributions

This work was carried out in collaboration between all authors. Authors OGF and OKO wrote the protocol and were both involved in the sample collection. Author BO carried out the SEM/EDX and optical microscopy analysis. Author OGF carried out the air-mass trajectory analysis and the draft of the initial manuscript. All authors were involved in the final compilation and editing of the manuscript.

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Original Research Article

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ABSTRACT

The prevalent problem of air pollution in Nigeria is attributable to the country's huge population, put in excess 160 million and the trend of industrial growth. In 2010, the country witnessed a rare occurrence of 9-day period of heavy dust episode (HDE). The dust reduced visibility to < 1 km, causing cancellation of several flights. The aim of this study is to assess the level of aerosol mass loadings and, nature and sources of the HDE aerosols. Two fractions of particulate matter (PM) were collected for about two months including the period of the HDE. Samples were collected on Whatman polycarbonate filters using low-volume GENT sampler equipped with a stacked filter unit (SFU) to hold two 47 mm filters. 7-day back trajectory analysis was performed using UGAMP trajectory model driven by ECMWF wind analyses data. Morphological analysis of the PM was

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done using Scanning Electron Microscopy (SEM), chemical compositions determined with Energy Dispersive X-ray (EDX) and particle number per unit area of filter (particle density) was estimated using optical microscopy. The range of mass concentration of $PM_{2.5}$, $PM_{2.5-10}$ and PM_{10} are 1.24 – 58.7, 5.1 – 354.9 and 8.33 – 379.2 μg m^{-3}, respectively. EDX detected twelve (12) elements: Fe, Na, Mg, Al, Si, S, K, P, Cl, Ca, Mo and O. Elements of crustal origin (Si, Al, Fe, Ca and Mg) account for a high percentage of the elemental composition of the PM. Four distinct classes of particles - mineral dust, NaCl containing agglomerate, Calcium-rich dust and alumina-silicate - were identified from the morphological and compositional analysis of the PM. From the backward trajectory analyses, most of the crustal components of the HDE dust can be attributed to long-range dust transport from North Africa and the Sahel region, while maritime aerosols are attributable to the marine environment in the Atlantic, down South.

Keywords: Dust episode; Nigeria; SEM/EDX; air-mass trajectory; optical microscopy.

1. INTRODUCTION

Airborne Particulate Matter (PM) are pollutants emitted into the ambient air in condensed form; solid and/or gaseous phase [1]. They are emitted into the atmosphere from either anthropogenic or natural sources [2-4]. Their impacts on human, the environment, animal and plant have been of major concern to every stakeholder. Extensive studies have been carried out to provide adequate understanding of the nature, sources and impact of PM. Elevated PM concentrations have been linked to high morbidity and mortality rate, increased hospital admission for respiratory related illnesses, chronic respiratory and heart diseases [5-12]. PM have also been identified as a major contributor to climate forcing [13-16], reduced crop yield [17] and damage of vegetation [18-20].

The intensity of these impacts on human, vegetation and the environment varies with the mass concentration, particle size, morphology and the elemental composition of the PM [18]. Emission from vehicles, industrial processes, domestic heating and cooking and, traffic related (mainly from tyres and brake lining) have been identified as major anthropogenic sources of PM [21,22]. Identified natural sources are re-suspended dust, sea spray, volcanic eruption and long-range dust transport [4,23]. Adequate knowledge of their chemical composition and structure (morphology) will enhance a better understanding of their detrimental effects on human health and a possible pointer to their sources [24,25].

Dust episodes enable long-range transport of disease causing pathogens - bacteria, virus and fungi - even across hemispheres [26-28]. At various places around the globe, increased cases of conjunctivitis, rhinitis [29,30] and, allergic and non-allergic respiratory illnesses – asthma [31], silicosis/pulmonary fibrosis, severe cough and respiratory tract infection [32,33] - have been attributed to desert dust which is often associated with elevated level on bacteria, fungi and virus in ambient air during and after the dust episode [34-36].

Trajectories have been defined as the paths of infinitesimally small particles of air [37]. Back trajectories analysis has been a useful tool to track and understand the history of air parcel arriving at a location of in situ measurements [38,39], remote measurements [40], and measurement platforms on flights [41,42]. At the measurement locations, sources contributing to aerosol measurements can be identified and further investigated using the history of air mass arriving at such locations.

The Nigerian weather has two predominant seasons; the rainy (May – October) and dry season (November-April). The month of March is usually the peak of the dry season. Heavy dust episodes (HDE) are very rare occurrence in Nigeria especially in the South-western part of the country. As such, the HDE experienced between March 17 and 26, 2010 came as a huge surprise. Its intensity peaked between the 21st and 25th, reducing visibility significantly to less than a kilometer causing the cancellation of several flights.

To the best of our knowledge, there has not been a study on the analysis of the composition, morphology and sources of aerosols from dust episodes in Nigeria. As such, this study aim to provide a pioneering work in this regards. This study explores an analysis of the air-mass history and composition - morphology, particle density (particle number) and elemental - of the HDE aerosols. This will enable an assessment of the

contribution of the two fractions – fine and coarse - sampled, elemental composition of aerosol loadings and identification of the possible major origin(s) of these aerosols.

2. MATERIALS AND METHODS

2.1 Sample Collection

Samples of PM2.5 (fine) and PM2.5-10 (coarse) were collected on nuclepore polycarbonate filters using a low volume Gent sampler [43,44]. The sampler was equipped with a stacked filter unit (SFU) to hold two 47 mm filters of 8.0 and 0.4 µm pore size. The filter holder was placed facing downwards, at a height of about 1.6 m, to avoid passive settling of particles on the filters. Polycarbonate filters are best suited for the microscopic analysis of PM [45]. Ambient air was sampled at an average rate of 16 L/min on a 24-hour basis. The sampling was carried out at the top of the two-storey Physics building of the Obafemi Awolowo University, Osun state, Nigeria (7.52℃N, 4.52℃E and 294 m above sea level). The maps in Fig. 1 show the location of Nigeria (study site) in relation to North Africa and the Sahel region - the dominant source of dust on the Africa continent. It also shows the map of Nigeria and a blown-up Google map showing the sampling location within Nigeria. Samples with identification number 42, 46, 47, 50 and 54 (fine and coarse) were all collected between March 9 and 31, 2010, both days inclusive. Samples 46, 47 and 50 were collected during the peak days of the HDE.

The filters were humidity conditioned in desiccators before and after sampling and allowed access to ambient air in the laboratory so as to equilibrate with ambient air condition. To avoid contamination of the aerosol samples, the filters were handle with a sterilized forceps and were kept in well labelled sample dish. Pre- and post-sampling weights of the filters were determined using an electric microbalance (Sartorius model CP2P-F). The difference in the weights - pre and post-sampling - gives the mass concentration of the PM on the filters, measured in µg m^{-3}.

2.2 SEM/EDX Measurement

Scanning Electron Microscopy (SEM) uses a focused beam of high-energy electrons to generate a variety of signals at the surface of solid specimens. The signals that derive from electron-sample interaction reveal information about the sample including external morphology (texture), chemical composition, and crystalline structure and orientation of materials making up the sample. The morphology and elemental analysis of the aerosol samples were done using Hitachi S-4700-II Scanning Electron Microscope with Energy Disperse X-ray Spectroscopy (EDX) facility attached [45-47]. The filters were mounted on the SEM stub with adhesive carbon tape. For each of the filters, secondary electrons were acquired to produce the SEM micrograph images at different magnification under 10 kV accelerating voltage with a working distance of 10 mm.

For the EDX analysis, different particles from the samples were selected and analysed for elemental composition. X-ray spectra were collected with an accelerating voltage of 20 kV and emission current of 21 kA. The EDX counting time was 20 s lifetime for each particle at magnification ranging between 3000x and 10000x. The net x-ray intensities of various elements were obtained and the spectrum plotted [48]. These intensities were also converted to weight fractions and atomic fractions using EDX software with ZAF (Z is the atomic number, A, the absorbance and F, the fluorescence values of the element) quantification. The EDX spectra were analysed to determine the relative abundance of 12 elements - Al, Si, S, Cl, Na, Mg, Ca, Fe, K, P, O and Mo.

2.3 Optical Microscopy

The optical microscope, often referred to as the 'light microscope', is a type of microscope which uses visible light and a system of lenses to magnify images of small samples. Olympus BH2 optical microscope with top illumination approach was used for the microscopic analysis of particles on the filters. Four different areas were photographed and analysed on each filter. Each of the micrograph was analysed using Olympus Stream Essentials 1.7 (Olympus Corporation). The average area covered during each shot on the filters was 2 x 10^5 μm^2. Table 1 show the particles number in each photographed area (particle density) on the filters.

2.4 Trajectory Analysis

With a view to investigate the history of air mass arriving at the sampling site, 7-day backward trajectory analysis was undertaken. The

trajectories were calculated using the UK Universities Global Atmospheric Modeling Programme (UGAMP) trajectory model. The model is described extensively elsewhere [49]. The model is driven by the 6-hourly European Centre for Medium-Range Weather Forecast (ECMWF) wind analysis data. The UGAMP which uses the fourth-order Runge-Kunta integration method, is a well-tested, widely used and validated offline trajectory model [50]. It has been used in several in-situ and air flight measurement campaigns to study contributions of prevailing air-mass trajectories to measurements [51-55]. Twenty-five (25) trajectories are released at 12:00 UTC from the sampling site at an atmospheric pressure of 900 hPa on each of the days investigated. The back-trajectories contain information output at time-steps of 0.6 hours on each of the clusters.

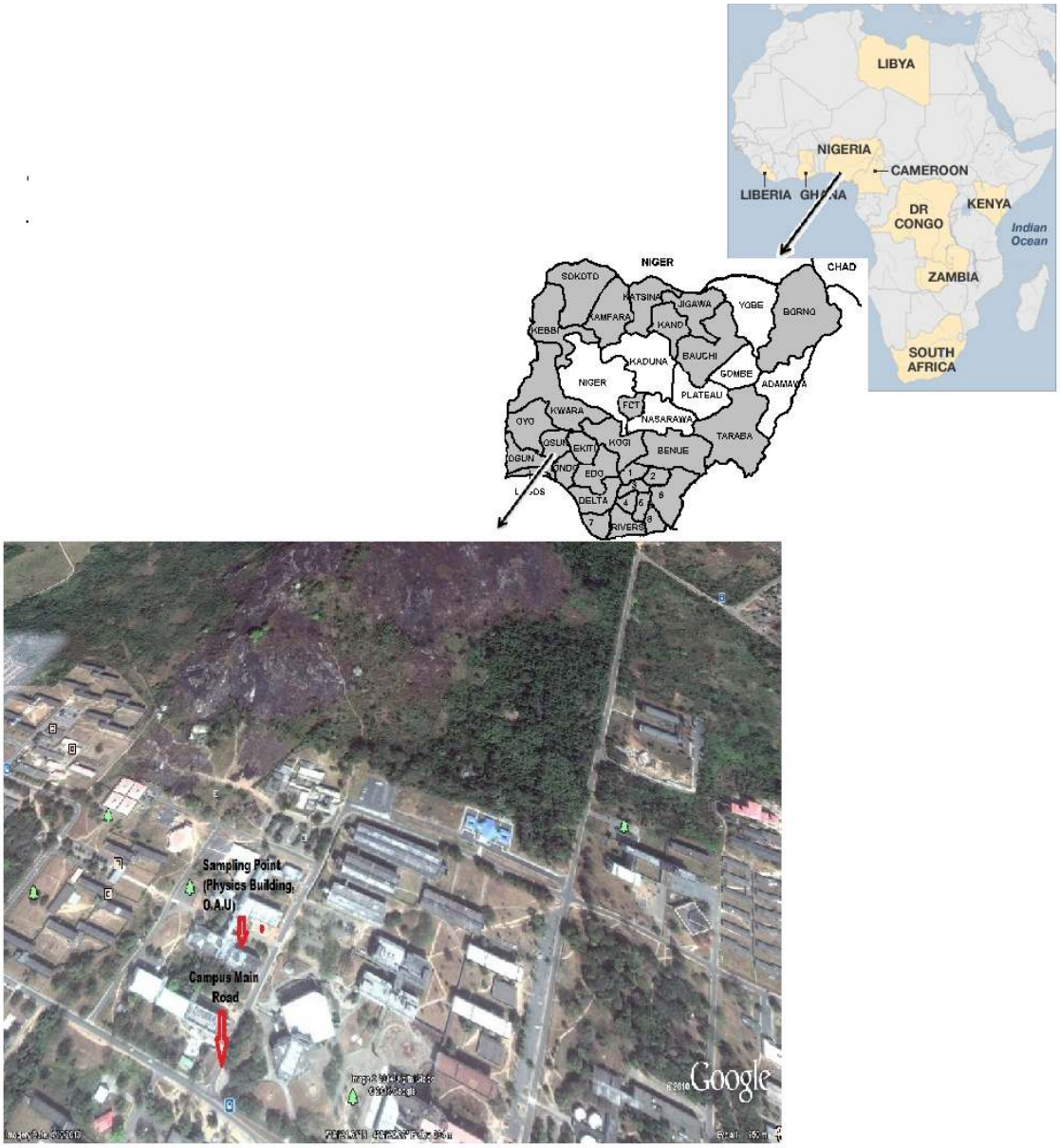


Fig. 1. Map showing the location of Nigeria on the map of Africa and blown-up Google map showing the sampling site

3. RESULTS AND DISCUSSION

3.1 Mass Concentration of PM

During the period of the HDE, the mass concentrations of the coarse (PM2.5-10) and fine (PM2.5) fractions were about 8- and 4-folds of the non-HDE period, respectively, as shown in Fig. 2. The highly bloated mass concentration of the coarse fraction underpins the fact that a large proportion of the HDE PM is due to long-range dust transport rather than vehicular emission or open burning processes that had been identified by previous studies as the prominent sources of PM in Nigeria [4,56]. The PM mass concentrations obtained for PM2.5 and PM10 during the HDE period ranged between 1.24 – 58.7 and 8.33 – 379.2 μgm^{-3} respectively. These values are 2- and 7-folds of the World Health Organization's limits of 25 and 50 μgm^{-3} for PM2.5 and PM10 respectively. Samples 46, 47 and 50 were collected during the peak period of the HDE. The particle counts on the coarse fraction of these samples (46, 47 and 50) are higher compared to samples 42 and 54, which are pre- and post-HDE samples respectively (see Table 1). In Table 1, the areas represented as I – IV are portions of approximately 2 x 10^5 square micrometres on some filters of the HDE aerosols. Fig. 3 shows images obtained from optical microscopy analysis of individual particles in the area photographed on some filter samples of the HDE aerosol. Figs. 3a and b are coarse fraction while c and d are fine PM fraction. As shown from the microscopy images and PM mass concentration, the HDE aerosols are mainly in the coarse fraction. This can be attributed to long-range dust transport from the Sahara during the HDE period.

3.2 Elemental Distribution and Particle Morphology

Three sets of particle photomicrographs (SEM and optical microscopy) and EDX elemental spectra were obtained for each of the ten (10) filter samples. The EDX ZAF quantification of the elements on the HDE coarse and fine filters (46, 47 and 50) shows higher concentrations for crustal elements (Al, Si, Mg, Ca, Fe), especially for the coarse PM fraction. There were trace concentrations of open burning signature elements (P, K, S, and Mo) which might have been picked up as the air masses travel between the source and receptor sites. Table 2 gives the elemental concentrations obtained from EDX

Table 1. Particle count (PC) in the photographed areas on the filters

Sample Id	Date	Area I	Area II	Area III	Area IV	Average PC
C42	09/03/2010	196	189	187	187	189.8
C46	17/03/2010	363	309	309	313	323.5
C47	19/03/2010	299	309	360	360	332.0
C50	23/03/2010	257	257	268	268	262.5
C54	31/03/2010	176	190	164	196	181.5
F42	09/03/2010	235	247	307	300	272.3
F46	17/03/2010	330	269	237	288	281.0
F47	19/03/2010	451	469	526	513	489.8
F50	23/03/2010	101	96	112	93	100.5
F54	31/03/2010	331	297	307	348	320.8

Table 2. Elemental concentration of PM samples (% weight)

	Al	Si	S	Cl	Na	Mg	Ca	Fe	K	P	Mo	O	Date
C42	0.31	0.23	-	2.25	4.53	0.16	4.99	-	0.24	-	-	19.74	09/03/2010
C46	1.69	6.54	0.3	0.21	0.97	0.43	2.37	0.82	0.61	0.7	-	26.33	17/03/2010
C47	0.30	1.27	0.72	-	0.05	-	9.53	-	0.77	-	-	22.58	19/03/2010
C50	11.04	13.2	-	0.22	0.31	0.1	1.97	5.07	1.38	-	0.44	30.85	23/03/2010
C54	3.36	5.73	0.13	0.21	0.86	-	-	1.94	0.87	0.23	-	25.6	31/03/2010
F42	0.21	0.05	1.99	0.7	1.53	-	5.18	-	1.14	-	-	14.38	09/03/2010
F46	3.15	9.67	0.71	0.51	0.78	0.15	1.1	1.86	2.21	-	-	25.15	17/03/2010
F47	5.39	9.06	-	-	0.23	0.06	2.05	-	0.7	-	0.4	27.36	19/03/2010
F50	7.03	7.35	-	-	-	-	0.32	2.98	0.32	-	-	30.81	23/03/2010
F54	2.2	7.35	2.57	0.2	0.94	0.08	-	1.8	2.53	-	-	20.4	31/03/2010

analysis for the samples, as percentage weight. Carbon was not used for the classification of the particles, as it was difficult to quantify the contribution of the polycarbonate filter to the carbon abundance. The particles classification was done taking into consideration the particle's elemental composition and morphology. The identified classifications are mineral dust, NaCl containing agglomerate, calcium-rich dust and alumino-silicate.

3.2.1 Mineral dust particles

These particles have a high concentration of silicon (~41.6 %), which is presumed to be made up of SiO_2 (such as quartz). Aluminium, iron, sulphur, magnesium, potassium and sodium make up the remaining ~58.4%. They are predominantly crustal elements which must have been due to soil re-suspension and largely to long-range dust transport from North Africa and the Sahel region. This is evident in the elevated concentration of $PM_{2.5-10}$ during the period of HDE. Mineral dusts constitute a huge chunk of atmospheric global aerosol and are *aeolian* in nature [57-59]. The SEM/EDX spectra and micrographs of particles in this class are shown in Figs. 4a and b. This class of particles dominate the samples collected during the peak periods of the HDE.

3.2.2 NaCl containing agglomerate

Particles in this classification contain mainly sodium and chloride, with trace amount of calcium, potassium and molybdenum. In Nigeria, studies have attributed NaCl to contribution from sea-salt [4,21]. This class of particles are found in higher concentrations, in both the fine and coarse fractions, on the pre- and post-HDE samples. The morphology of this class of particle shows an almost cubic shape often attributed to NaCl particles (Fig. 4c). The slight deformation of the cube-shaped image might be due to the presence of Ca and Mo as 'impurities'. Molybdenum in ambient air PM loading is a maker element for the iron and steel smelting industry [60].

3.2.3 Calcium-rich dust

Calcium strongly dominants this class, making up for about 75% of the elemental concentration of the class. Contributions from silicon, sulphur and potassium make up the remainder of the class. Some of these particles are probably limestone ($CaCO_3$) which is from dust (clay) and industrial processes. The SEM micrograph showing the morphology and the EDX elemental spectra of the particles in this class are shown in Fig. 4d.

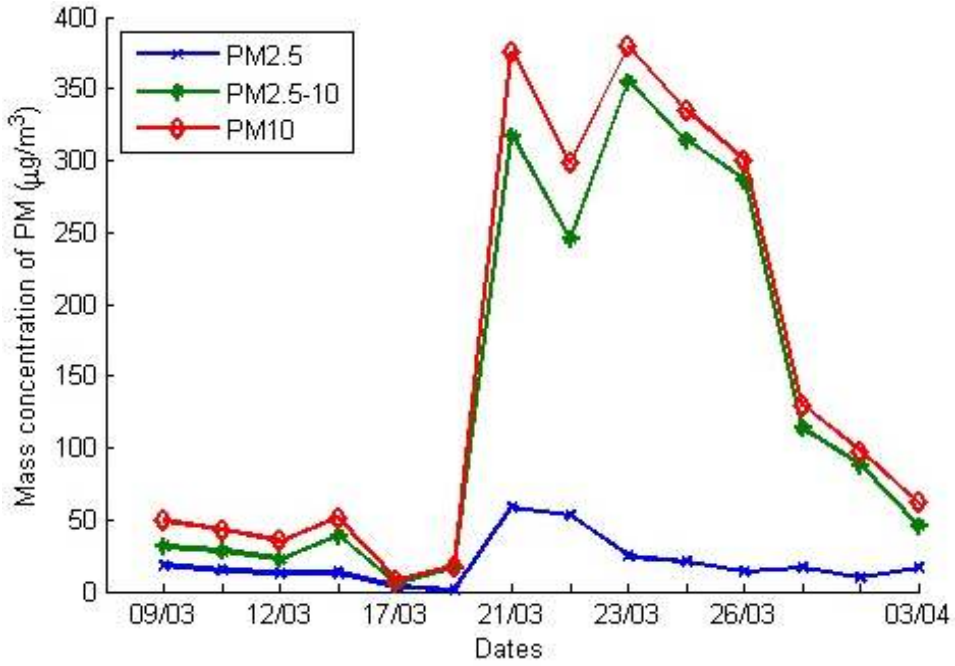


Fig. 2. Time series plot of the mass concentration of PM fractions before, during and after the HDE

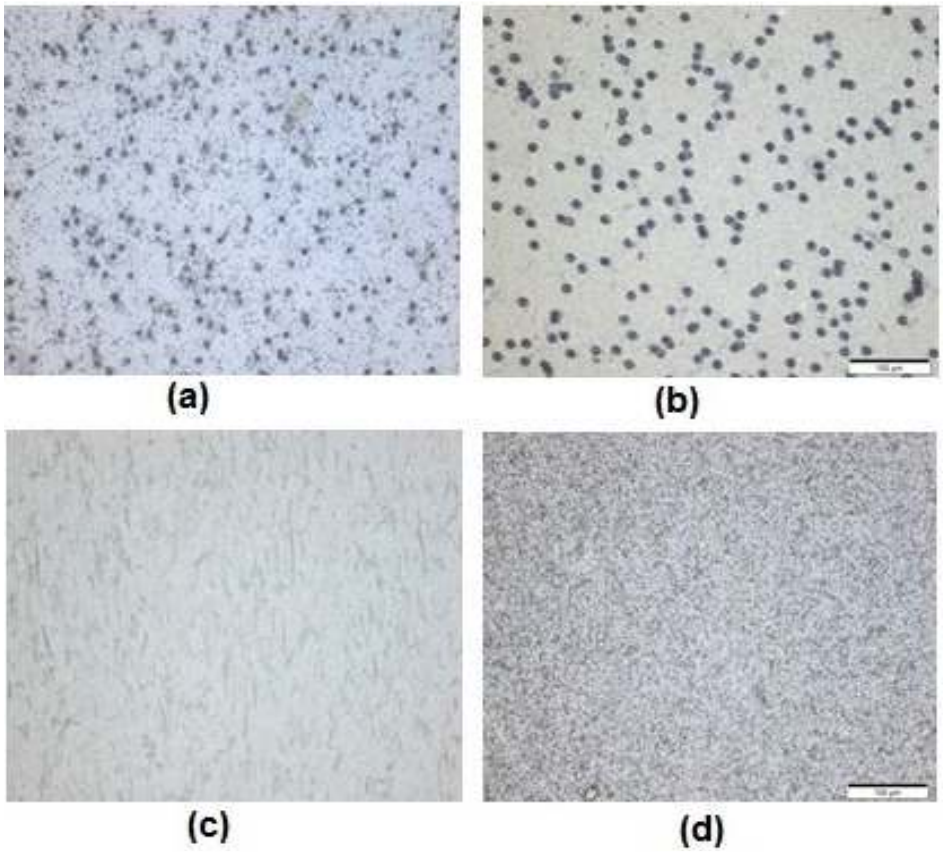


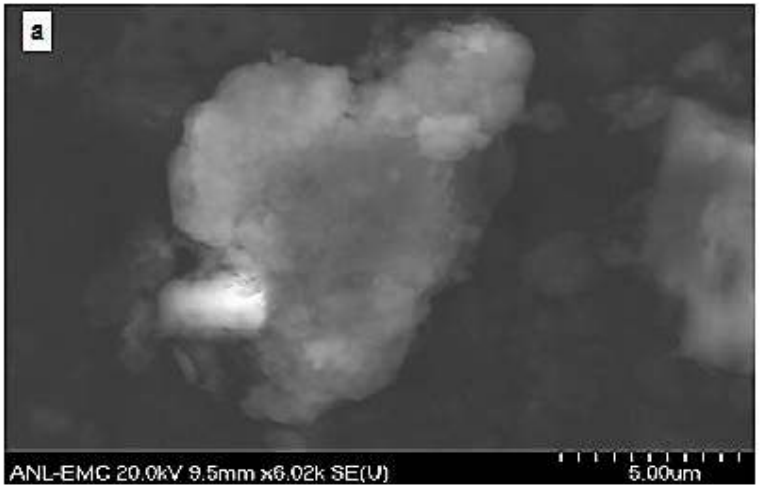
Fig. 3. Optical micrograph obtained from optical microscopy showing particle counts on photographed areas on some HDE filter samples

3.2.4 Alumino-silicate

Aluminium and silicon together account for over 90 % of the elemental concentration of particles in this class with trace contribution from iron. The SEM micrograph and x-ray spectra are shown in Fig. 4e. The major sources of this class of particles are wind-blown rock and soil particles [61]. This underpins the fact that aerosols in the HDE are predominantly *aeolian* soil dust due to long-range dust transportation which is responsible for the bloated mass concentration of PM during the HDE period at the study site.

3.3 Back Trajectory Analysis

Figs. 5(a – g) show the trajectory plot for periods before (March 12 and 17), during (March 19, 22, 25 and 26) and after (March 31) the HDE. Fig. 5h shows the colour coding of the atmospheric pressure, in hPa, as the air masses travel from the source to the sampling location. Plots 5a and b show that pre-HDE aerosols are contributed, predominantly, by air masses from the South and South-east over the Atlantic. Aerosols sampled during this period show elevated concentration of maritime aerosol as indicated by the Na and Cl concentrations. Plots 5c, d, e and f of air masses trajectories during the HDE period, show air masses from North Africa and the Sahel region as the major contributor to the aerosol sampled during period. These plots (5c, d, e and f) show that the bloated mass concentration of the coarse fraction experienced during the HDE are due to long-range transport from North Africa and the Sahel region. Plot 5g shows a return of the dominant contributing source to the South maritime area after the HDE period. This trajectories analysis shows that a sufficient number of the air masses from the source of the aerosol to the sampling location during the HDE period are within 900 and 1000 hPa - the planetary boundary layer (PBL) suggesting that the HDE aerosols are within the areas of direct contact with human and as such can be easily inhaled at high 'doses'.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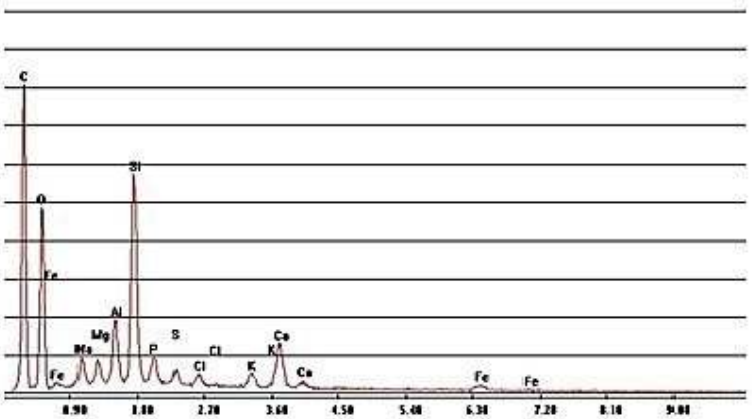
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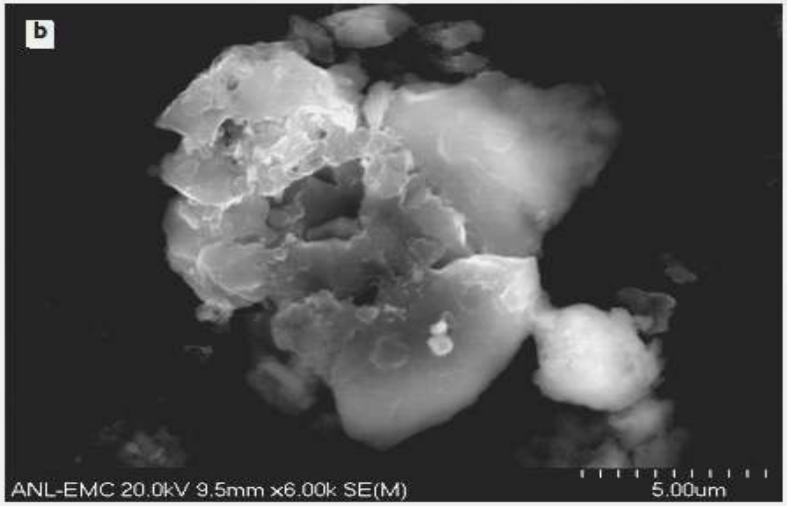
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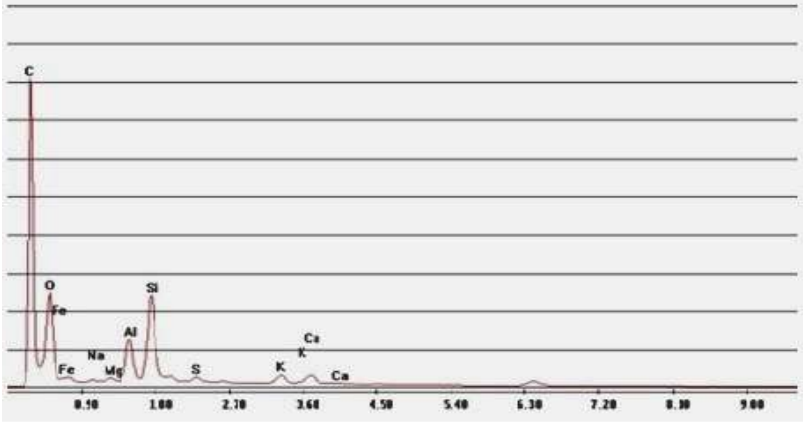
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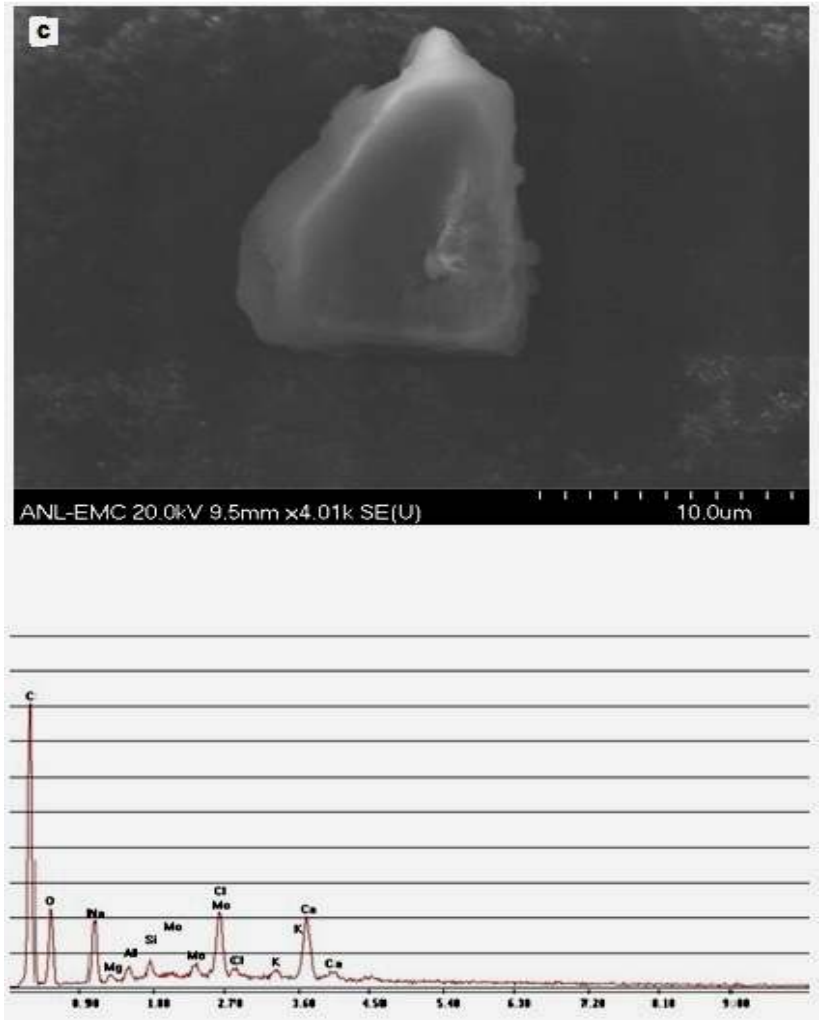
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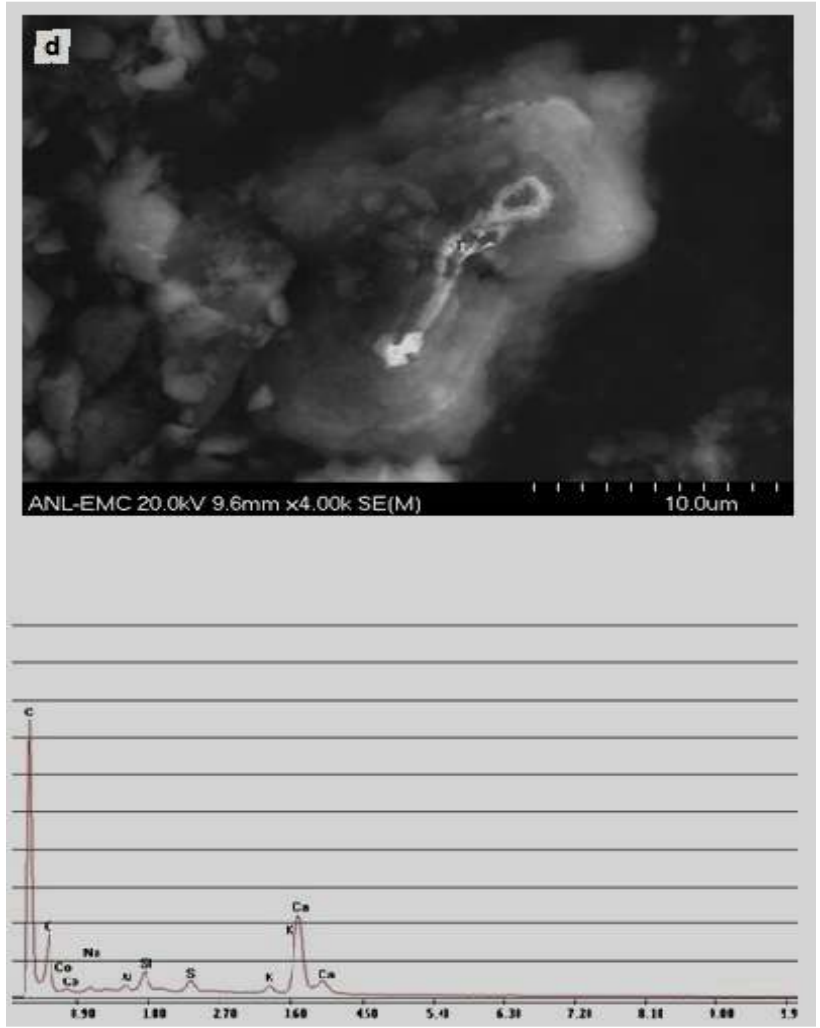
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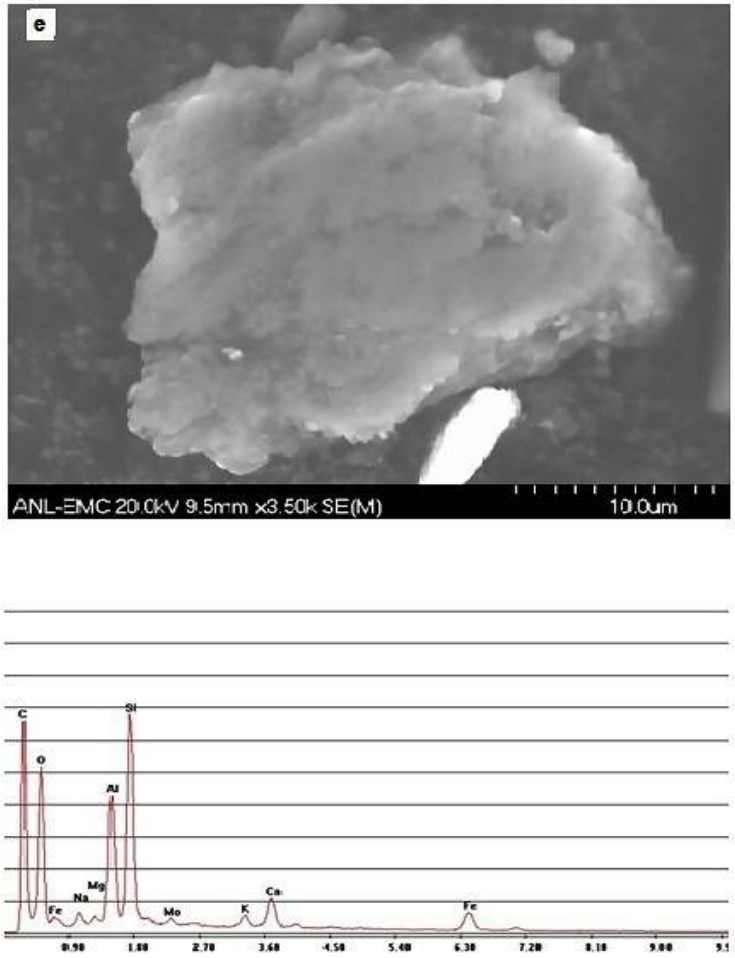

Fig. 4. Morphology and elemental X-ray spectral from EDX for selected particulates from HDE aerosol samples (a and b: mineral dust particles; c: NaCl agglomerate; d: Ca-rich dust and e: Alumino-silicate)

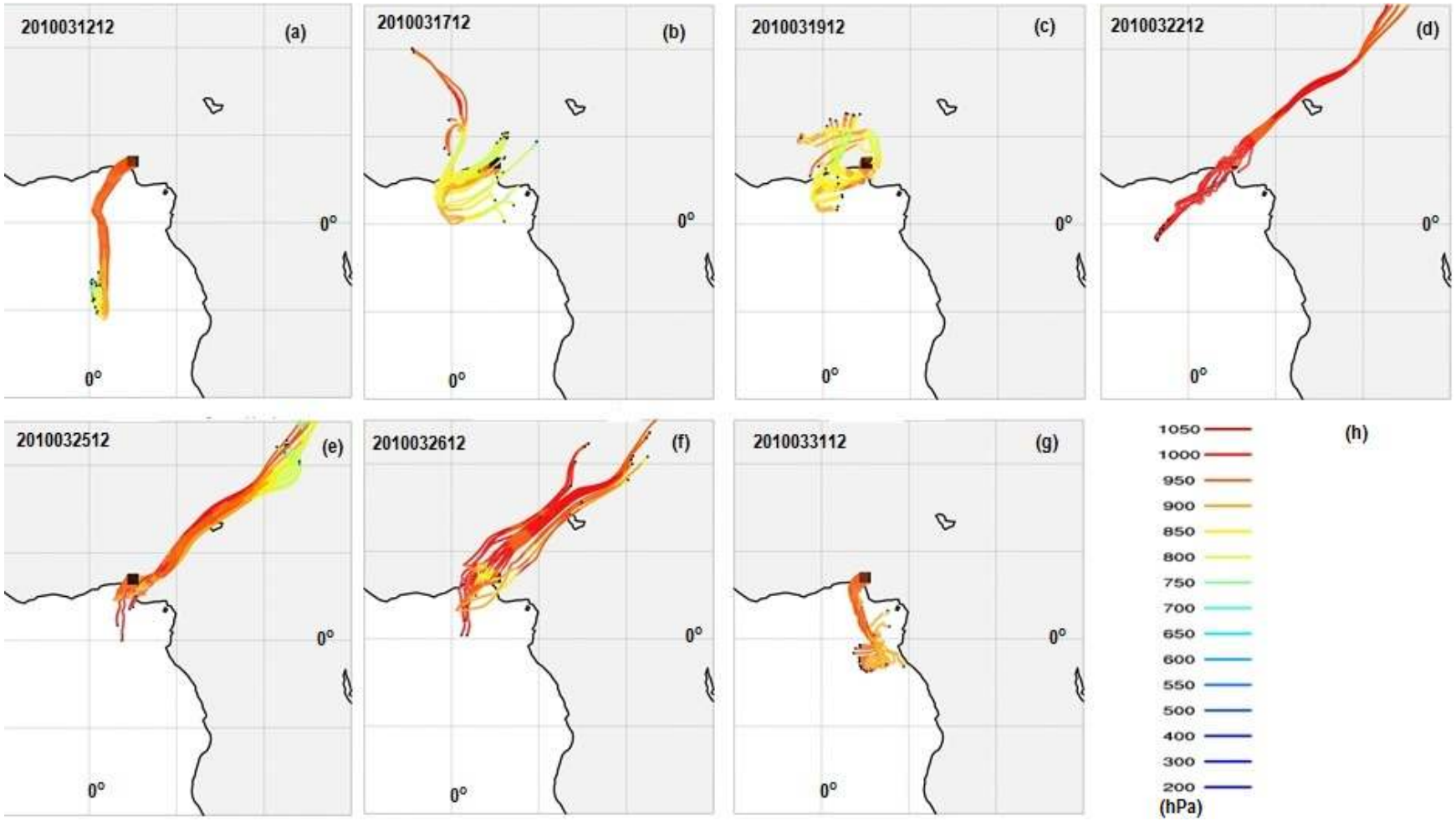

Fig. 5. 7-day backward trajectory analysis for the pre-, post- and HDE periods

4. CONCLUSION

This pioneering work on the analysis of the composition, morphology and origin of dust episode aerosols in Nigeria identified highly elevated mass concentration of two fractions of PM: ~ 8- and 4-folds of non-HDE periods for fine and coarse fractions, respectively. Element of crustal origin (Al, Ca, Mg, Si, Fe) are of higher concentrations in the HDE PM while those of maritime origin (Na and Cl) have elevated concentration in the pre – and post-HDE samples. The study identified four classes of particle in the samples, namely, mineral dust, NaCl containing agglomerate, calcium-rich dust and alumina-silicate, majority of which were due to soil re-suspension and long-range dust transport. Air-mass trajectory analyses show that maritime aerosols were contributed by air parcel originating from the South and South-east over the Atlantic while aerosols of crustal origin were contributed by air masses emanating from North Africa and the Sahel region. As shown in the trajectory plots, air masses travelled within the PBL (atmospheric pressure > 950hPa), through Nigeria cities and town, from the source region to the receptor site. This suggests likely inhalation of the dust in high 'doses' by man and animal.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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