

Contribution of Kerosene Soot Particulates to Domestic Emissions

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Abstract:

One question, uppermost in combustion study is whether soot samples emit toxic particulates and could contribute to air pollution, occupational and domestic hazard on exposure of the populace. In this study, we document experimental report on the protocols and levels of pollutants emitted from soot into the near atmosphere. Characterization of soot was carried out using X-Ray Fluorescence (XRF) spectrometry, X-Ray diffraction (XRD), Scanning Electron Microscopy (SEM), and Fourier Transform Infra-red (FTIR) Spectroscopy. The pH of soot samples is near the neutral range (6.60-6.85). Samples are majorly covalent as revealed by the zero conductivity values. The moisture contents of KLS (10.0%) is almost twice, as was reported for KSS and are typical to explain the tendency of the soot dusts to form aerosols in near atmosphere. Poor or low values of bulk densities (0.058-0.060 g/cm³) is an indication that the soot can be easily spread by air current to the environment. Levels of impure carbon revealed by EDS analysis clearly pointed at the organic pollutants loading potentials of the soot. Other findings include FTIR characteristics signals indicating aromatics, XRF analysis showing high concentration of Chromium and other toxic metals. Overall, impure carbon from kerosene based soot could contribute to local emissions, mostly produced by the sources with low emission height. Secondly, the levels of toxic metal in soot samples exceed the permissible legislative limit for air samples.

Keywords:

Air Pollution, Aerosols, Kerosene, Soot, Carbon, Metals, Particulates

1. Introduction

Combustion has had significant impact on our daily life since the beginning of human history. However, in addition to making our lives easier and better, combustion also negatively affects our society. It threatens human lives by generating environmental

pollutant such as oxides of nitrogen (NO_x), and by changing the global climate pattern via greenhouse effects. Dust and soot fragments forms components of air particulates, which are released into the air as extremely small particles or liquid droplets [1]. Several reviews pointed at the challenges of air pollution. One among them is Particulate Matter Pollution [2].

The main source of environmental soot is the combustion of fossil-based fuels and biomass burning at the Earth's surface [3]. The other examples of soot may include coal, charred wood, petroleum coke, chemosphere, and tars[4]. Soots are considered as unwanted byproducts derived from incomplete combustion of carbon-containing materials [5]. Soot is a powdery mass of fine black particles [6]. It consists of impure carbon, formed after the incomplete combustion of hydrocarbons [7]. To some extent, settled dust, cooking, oil lamps, smoking of plant matter, fireplaces, candles, house fires, furnaces, and local field burning also contribute to the soot production [8]. The relative amount of elemental carbon inside soot is considered to be less than 60% of the total mass of particle [9]. Among hydrocarbons, the poly aromatic hydrocarbons (PAHs) are the main carcinogenic compound in the soot [10].

The impact of soot on the human health and to the entire environment depends on its distribution and its distance from the source of origin [11]. It has been believed that the vehicle exhaust contributes to approximately 50% of urban particulate matter (PM) [12]. The special attention is given to the smaller fractions of PM (PM 2.5 and PM 0.1) because these particles can penetrate deep into the bronchiolar parts of the lungs and cause various health hazards [13]. The smaller the particles, the deeper they permeate the lung and the more harmful they are for individual's health, especially the ultrafine particles under 2.5µm which can reach the alveoli [14].

Kerosene is the major refinery product before the onset of the automobile age [15]. However, the conversion of energy from one form to another often affects the environment and the air we breathe in many ways, and thus the study of energy is not complete with-out considering its impact on the environment [16].

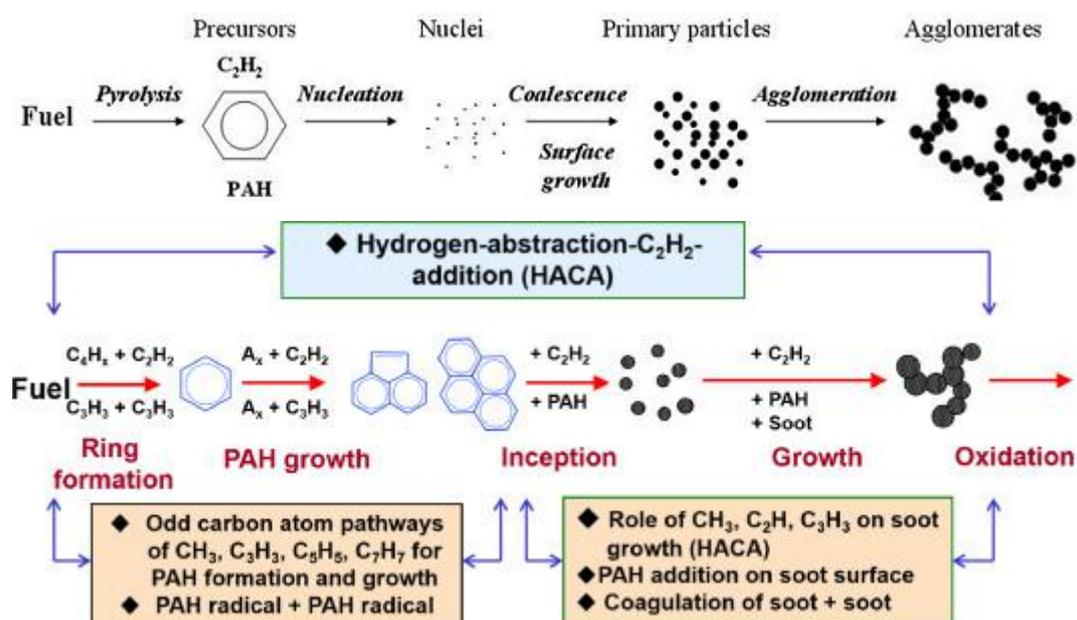


Figure 1. Soot formation [17,18].

Soot Formation Mechanisms: The evolution from liquid or vapour phase hydrocarbons to solid soot particles involves five processes (Figure 1): pyrolysis,

nucleation, coalescence/surface growth, agglomeration and oxidation. Only the oxidation can convert hydrocarbons to CO, CO₂ and H₂O at any time in the sequence.

Particulate matter: The contribution of carbonaceous matter to the total particle composition cannot be over emphasized due to its effects on health, visibility, and climate. Steven et al. [18] reported that carbonaceous matter contribute significantly to the global sub-micrometer particle emissions

Health Impact: Particles emitted from combustion sources are harmful to human health and affect radioactive balance in the atmosphere. Aerosol particles affect air quality locally and regionally but have global influences as well. Adverse health effects are a concern especially when considering local and regional particle emissions. Local emissions are mostly produced by the sources with low emission height such as traffic and small-scale combustion. These emissions can also impact regionally. Instead, large-scale combustion of fossil fuels where emission height is high mainly has regional influence. The health effects of particles are influenced by local, regional and long-range transported emissions but the climate effects are global due to intercontinental transport of polluted air masses. Soot penetrates deep down in the lungs, imparting a wide range of serious health outcomes, including acute bronchitis and aggravated asthma among children, heart attacks, and strokes, as well as premature death [19]. Further, the American Lung Association [20] opined that inhaling soot can potentially cause “cancer as well as developmental and reproductive disorders”.

Carcinogenicity of soot: The routes of entry into the human body include ambient and indoor air inhalation, ingestion of contaminated food, and dermal contact. Based on sufficient evidence from human carcinogenicity studies, soot causes cancers in humans [20]. In 1775, British surgeon Percivall Pott established a correlation between scrotal cancer and exposed chimney sweeps. In subsequent years, substantial amounts of epidemiological studies have since confirmed the elevated risk of scrotal and other skin cancers among chimney sweeps [21]. Correspondingly, follow-up studies among Swedish chimney sweeps revealed that risks for esophageal, hematopoietic, prostate, urinary bladder, and total lymphatic cancer were elevated [22].

Mutagenic/Genotoxic Effects: The mutagenicity of particulate matter is a function of its chemical composition and size. Pope et al. [23] carried out an analysis of the longer-term database of the American Cancer Society and found that long-term exposure to combustion-related fine PM is an important environmental risk factor for lung cancer mortality. A large amount of work examining the emissions from coal combustion has demonstrated the mutagenicity of both the polar and the aromatic fraction of the ambient particulate matter. Populations with a high occurrence of lung cancer have been linked to exposure with the PAH component of coal smoke.

This current research investigated the characteristics and chemical composition of two kerosene based soot samples from bush lamp (KLS) and kerosene stove (KSS), using classical and instrumental techniques. Soot mineralogy and the determination of pollutants (impure carbon and heavy metals) in the soot particles is the focus of this study.

2. Materials and Methods

2.1. Sampling and Sample pre-treatment

Sample collection method, documented by Atiku et al. [24] was adopted. Kerosene soot from bush lamp and stove was randomly scrapped from regions of high concentration on surfaces of their respective receptacles. 50 g of each sample was scrapped from the sources into clean sample holders with seals. They were labeled with KLS and KSS codifying kerosene lamp soot and kerosene stove soot respectively. Samples were stored at room temperature prior to analysis, using non-destructive analytical techniques. Analytical methods employed in this work include; Fourier Transform Infra-Red (Agilent tech. Cary 630), Energy Dispersive X-ray fluorescence Spectrometer (Mini PAL4), scanning Electron Microscopy (Q150R), and X-Ray Diffraction Instrument (X-SUPREME8000). Samples from the same sources were subjected to breaking of lumps to powder screening, harmonization and storage prior to analysis [24]. High analytical procedure was carefully followed throughout the experiment

2.2. Physiochemical Parameters

The physiochemical parameters of toner ink samples such as pH, conductivity, bulk density and moisture content were carried out using standard laboratory procedures. Important parameters are as follows:

The bulk density, using the tamping method [24].

$$\text{Bulk Density (g/cm}^3\text{)} = M_d/V \quad (1)$$

M_d = Mass of dry soot samples (grams), V = Soot volume

The moisture content, using the oven drying method [25].

$$M_{\text{soot}} = \frac{(W_i - W_f) \times 100\%}{W_i} \quad (2)$$

where; W_i = Initial mass of soot, W_f = Final mass of soot.

2.3. Characterization of Samples

Characterization methods documented by the National Metallurgical Development Centre; NMDC Jos, Nigeria was adopted with slight modification in mass- volume measurements. Elemental and chemical composition was based on XRF analysis. The Scanning Electron Microscopy (SEM) was used to examine the soot surface morphology, X-ray diffraction is for soot mineralogy and crystallographic parameters, including crystal sizes computed from peak positions at 2θ and X-ray counts (intensity), prescribed by the Debye-Scherrer's equation.

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (3)$$

where D is the Crysalline size, K is Scherrer Constant with value 0.9, λ is the wavelength of X-ray (1.540598), β is full width at half maximum and θ is the differential angle [26]. The functional groups present in the soot samples were detected on Fourier Transform Infrared Spectrometer.

3. Results and Discussion

3.1. Visual Inspection of Samples

Apparent colour of the kerosene based soot was examined with naked eye and found to be fine black particles, chiefly composed of carbon, they are extremely tiny about 2.5 micro meters or smaller in diameter. It is irritating to the eyes, nose and throat, and its odor may be nauseating. Due to its color and texture, soot tends to darken or stain surfaces. It stains ventilations, walls, ceiling, floors, clothes, and even skin. It is slightly sticky and tends to cling to surfaces [6]. Generally, soot is a powdery mass of fine black particles (plate 1).



Plate 1. Soot samples from (a) kerosene lamp and (b) kerosene stove.

a. Physiochemical Parameters

The physiochemical analysis of the kerosene based soot (Table 1) revealed the following;

pH – Results in Table 1 shows that the pH range for kerosene based soot and diesel based soot was between 3.34 – 6.85. According to definition, pH is a major of hydrogen ions concentration in a solution. This implies that a low pH values indicates a high hydrogen ions concentration. The pH values of the kerosene based soot and diesel based soot falls within the acidic range but the diesel based soot is more acidic than the kerosene based soot which is slightly acidic. The pH also determines the number of replaceable hydrogen in the carbonized samples which is an indication that metals are present in the samples.

Conductivity – Kerosene based soot has no conductivity; they produce no ions. Conductivity requires the existence of charged particles. This is an indication that the compound of the samples is mostly covalent with high carbon content [24]. Conductance is a measure of relative ease with which ions migrate. Therefore, the movement of this ions, i.e. positive ions (metals) will give a perception that heavy metals are present in the solution of the samples. It is well known that the conductivity of an electrolyte decreases as the solution is diluted while the molar conductivity increases upon dilution until a limiting value is reached [24].

Moisture Content- The results obtained in this analysis indicate the level of cluminess or ease of dissociation of the soot samples. The result in Table 1 shows moisture of a range between 5 – 20% was able to hold the soot as a whole bulk. The kerosene stove soot with less moisture can fragment or spread easily. Low moisture content could also be linked to high persistence and low microbial action (degradation) [27].

Bulk Density - Table 1 present the bulk densities for kerosene based soot, diesel based soot, printer ink and wood soot. Bulk density is a measures of strength and

attrition of the soot. All the values in this analysis shows values of less than 1. This is an indication that the soot can be easily spread by air current to the environment [24].

Table 1. *Physiochemical Parameters of Soot Samples.*

S/N	Parameters	KLS	KSS
1.	pH	6.60	6.85
2.	Conductivity ($\mu\text{f/cm}$)	0.00	0.00
3.	Moisture content (%)	10.0	5.0
4.	Bulk density (g/cm^3)	0.058	0.066

Table 2. *Elemental Composition (wt %) of KLS and SKS.*

Element Number	Element Name	Weight Conc. (%)	
		KLS	SKS
6	Carbon	80.75	82.38
8	Oxygen	17.41	15.61
7	Nitrogen	1.32	1.65
26	Iron	0.12	-
25	Manganese	0.10	0.04
16	Sulfur	0.10	0.10
13	Aluminum	0.08	0.08
14	Silicon	0.07	0.14
12	Magnesium	0.04	0.04

b. SEM Characterization of Soot Samples

The morphology and elemental composition (Table 2) of Kerosene based soot, was performed by the combination of different instruments. This is vital to obtain accurate surface morphology, mineralogical properties and compositions of each liberated particles in the soot and printer ink samples. The SEM micrograph of Kerosene based soot is presented. the surface morphology of the carbon deposit obtained is seen to be non-uniform. The kerosene soot particles are small. The SEM image of soot particles show particles of carbon which are chain-like agglomerations as shown in Plate 2. Energy dispersive spectroscopy (EDS) of kerosene soot is presented in Figure 2. The spectra show the presence of carbon and oxygen as the combustions product of kerosene. The composition of the soot aggregates from the EDS analysis of Kerosene lamp soot indicates the soot to consist of about 80.75% weight carbon and 17.41% weight of oxygen, 1.32% Nitrogen, 0.12% Iron, 0.10% sulfur, 0.08% Aluminium, 0.07% Silicon and 0.04 Magnesium were present. The kerosene stove soot also indicates the soot to consist of about 82.38% Carbon, 15.61% Oxygen, 1.65% Nitrogen, 0.14% Silicon, 0.10% Sulfur, 0.08% Aluminium and 0.04% Magnesium.

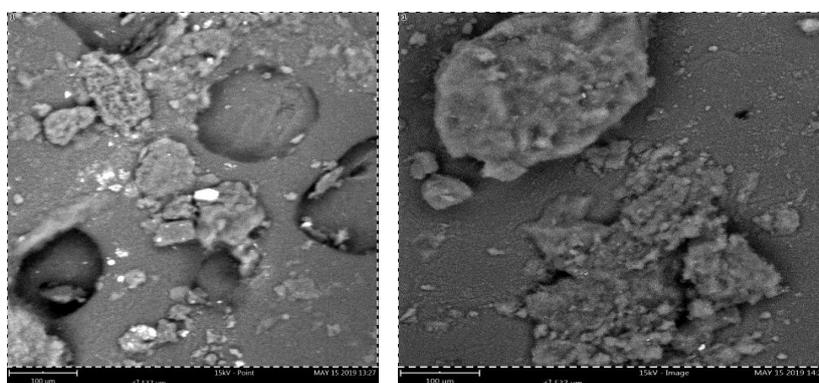


Plate 2. *SEM Image of (a) Kerosene Lamp and (b) Kerosene Stove Soot.*

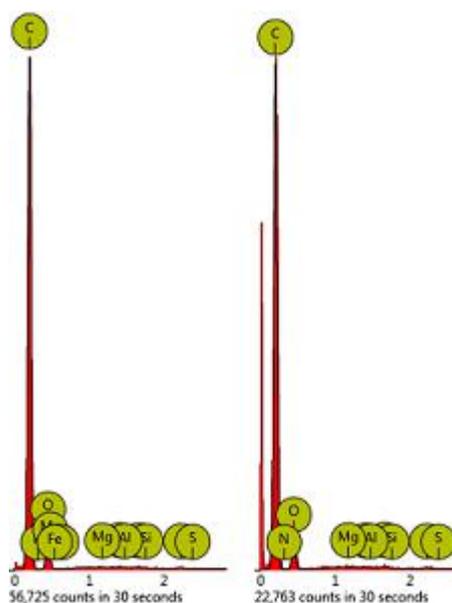


Figure 2. EDS Spectrum of (a) Kerosene Lamp Soot and (b) Kerosene Stove Soot.

c. FTIR Interpretation of Soot Samples and Printer Ink

The quantitative and functional groups of kerosene based soot samples were reported from FTIR technique. This chemical analysis revealed various spectral characteristics for each sample (Figure 3a and Figure b). For kerosene based soot the assignments of frequencies to their functional groups were carried out. The peak (Table 3) at 749.2 cm^{-1} for KLS, $7.45.5\text{ cm}^{-1}$ for KSS, and 738.0 cm^{-1} were attributed to aromatics. Aromatic hydrocarbons which contain more than one benzenoid (benzene-like) refers to as polycyclic aromatic hydrocarbons. Among major air pollutants representing health risks are polycyclic aromatic hydrocarbons, which can be found in the gas phase or well-bonded to particulate matter (PM) [28]. PAHs are a group of semi-volatile organic compounds composed of 2 or more aromatic rings, generated during incomplete combustion of organic matter. During this process molecules and radical fragment are combined, thus creating these substance [28]. These compounds are major environmental pollutants because they are considered to be potentially carcinogenic and mutagenic, hence considered “air quality markers” in terms of the health risks their presence represents [29]. The peak at 1581.5 cm^{-1} for KLS, 1581.8 cm^{-1} for KSS come mainly from primary amines. In the presence of light, amines released in the liquid or gaseous form react with atmospheric oxidants including oxidized Nitrogen compounds (photo-oxidation). The formation of nitrosamines is dangerous because these substances are toxic and carcinogenic even at very low concentrations [30].

Table 3. FTIR Spectral Information of Kerosene Based Soot Samples.

S/No.	Group Frequencies (cm^{-1})	Functional group	Observed Frequencies (cm^{-1})		Assignment
			KLS	KSS	
1.	2830-2695	Aldehydes	2765.7	-	H-C=O:C-H stretch
2.	2260.-2100	Alkynes	2109.7	2113.7	-C (triple bond)C-Stretch
3.	1760-1665	Carbonyl (general)	1703.4	1699.7	C=O stretch
4.	1650-1580	Primary amines	1581.5	1581.8	N-H bend

5.	900-675	Aromatics	749.2	745.5	C-H "OOP"
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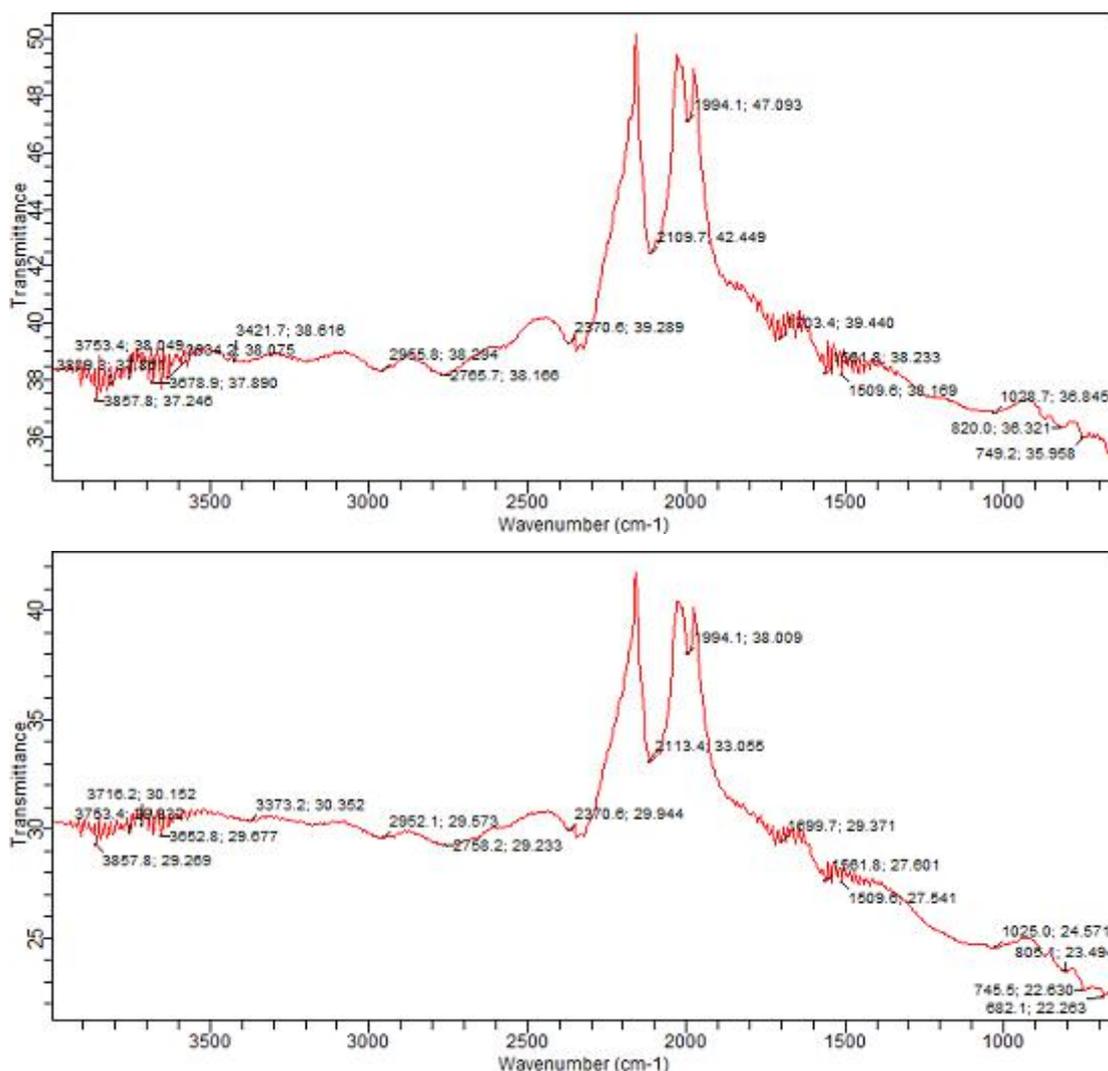


Figure 3. FTIR Spectrum of (a) KLS and (b) KSS.

d. XRF Characterization of Soot Samples and Printer Ink

Chemical characterization of the soot samples to investigate the elemental composition using energy dispersive X-ray fluorescence spectrometer (ED-XRF) revealed the results as presented in Table 4. The spectra for samples are presented in Figure 4.

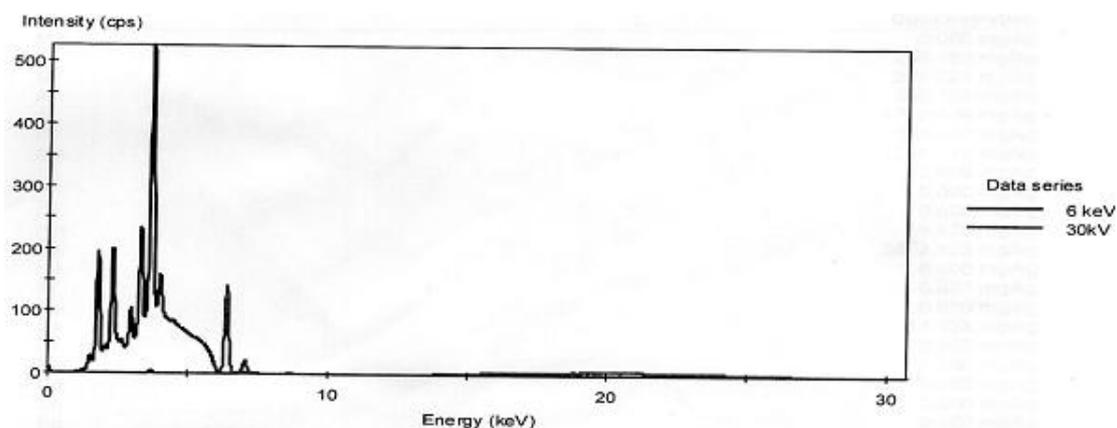


Figure 4. XRF Spectrum of Kerosene Lamp Soot.

The result revealed that kerosene lamp soot contains Mn 16.979, Zn 13.333, Ti 82.469, Fe 3672.492, Al 256.160, P 300.353, S 1775.174, Si 231.221, Cl 134.687, Ca 1299.712. Kerosene stove soot revealed the following; Mn 19.183, Cr 15.154, Ti 64.656, Fe 3050.349, Al 118.00, P 222.666, S 2478.544, Si 140.483, Cl 114.866, Ca 435.262 and wood soots unveiled levels of Mn 29.611, Cu 22.755, Zn 32.631, Ti 136.350, Fe 332.824, Pb 22.755, Sn 19440.988, Al 367.69, P 879.469, S 3478.185, Si 154.876, Cl 7374.882, Br 18.523, Ca 6717.887. Cu, Pb, Sn and Br are present in kerosene stove soot only. The presence of Chromium in the kerosene soot may be attributed to additives in the oil. Chromium occurs in the environment primarily in two valence states, trivalent chromium, Cr (III) and hexavalent chromium Cr (VI). Chromium (VI) is much more toxic than chromium (III) for both acute and chronic exposure [31]. Shortness of breath, coughing, and wheezing were reported in cases where an individual inhaled very high concentrations of chromium trioxide [32]. Chronic inhalation exposure to chromium (vi) in human results in effect on the respiratory tract, with perforation and ulcerations of the septum, bronchitis, decreased pulmonary function, pneumonia, asthma, and nasal itching and soreness reported [31]. Chronic human exposure to high levels of chromium (vi) by inhalation or oral exposure may produce effects on liver, kidney, gastro-intestinal and immune system and possibly the blood [32].

Table 4. XRF Elemental composition (mg/kg) of Kerosene Based Soot Samples.

S/N	Class	Element	Concentration (mg/kg)	
			KLS	KSS
1.	Heavy metals	manganese	16.979	19.183
		Chromium	ND	15.154
		Zinc	13.333	ND
		Titanium	82.469	64.656
		Iron	367.249	305.035
		Calcium	129.971	435.262
		Aluminum	256.160	118.000
2.	Non-metal	Phosphorus	300.353	222.666
		Sulphur	177.5174	2478.544
		Silicon	231.221	140.483
		Chlorine	134.687	114.866

Aluminium is also present in both Kerosene based soot and wood soot. Aluminium has been traditionally regarded as non-toxic metal that includes pathological changes only in very specific conditions e.g. long-term dialysis due to renal failure or occupational exposure to aluminium dust, fumes and its oxides [33]. Owing to the advanced knowledge and development of research methods, scientists have drawn their attention to aluminium assuming that it is an important etiopathogenetic agent responsible for the incidence of numerous degenerative diseases of the nervous system.

e. Mineralogy of Kerosene Based Soot (XRD Characterization)

The mineralogical component (Table 5; Figure 5.) of kerosene based soot samples were carried out by X-ray diffraction techniques. The main minerals found in the sample of kerosene stove soot is filipstadite. This compound have a phase information from the XRD pattern. the diffraction of X-rays by the crystalline solid sample results in a pattern of sharp Bragg reflections characteristics of the different d-spacing of the

solid. Broadening of these reflections beyond that due to instrumental factors is generally attributed to crystalline size effect [34].

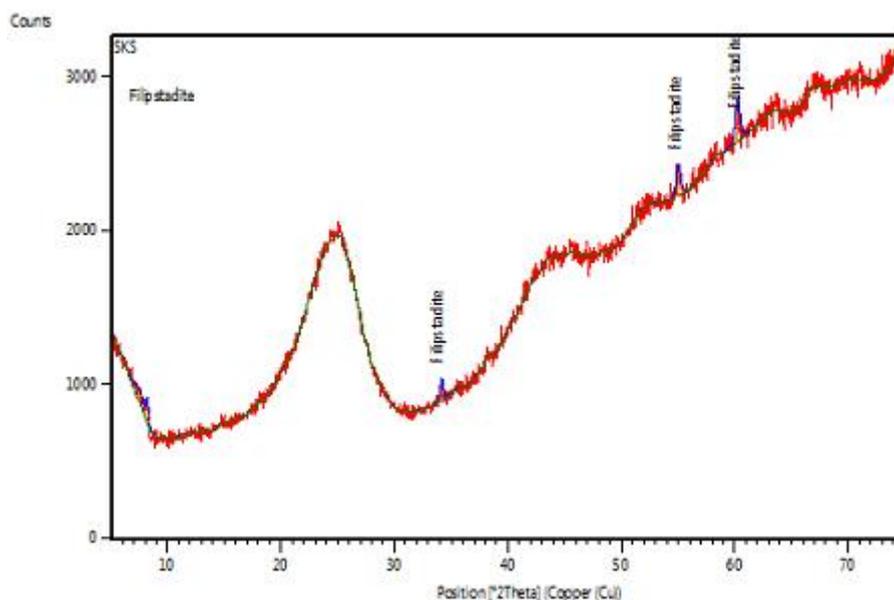


Figure 5. X-Ray Diffractogram of Kerosene Stove Soot.

Filipstadite in kerosene stove soot: This result reveals that the 2θ values at 29.212 and 34.398 show high intensities and this is attributed to crystallite size effects. Filipstadite is a very rare mineral of the spinel group, with the chemical formula $Mn_{10}Mg_6Fe_4Sb_4O_{32}$. It is isometric [35], although it was previously thought to be orthorhombic. When compared to a typical spinel, both the octahedral and tetrahedral sites are split due to cation ordering. Filipstadite is chemically close to melanostibite. The mineral comes from Longban, Sweden, a manganese skarn deposit famous for among rare minerals.

Table 5. Crystallographic Parameters of Kerosene Stove Soot.

Pos. [$^{\circ}2\theta$.]	FWHMLeft [$^{\circ}2\theta$.]	d-spacing [Å]	Crystalline size (nm)
7.4188	0.8187	11.91636	1.709
8.1897	0.3070	10.79622	4.560
34.1460	0.3070	2.62590	5.459
55.0090	0.4093	1.66935	5.900
60.2260	0.4093	1.53663	6.830
Average crystalline size			4.89

4. Conclusions

The chemical contents of soot samples were estimated. Low moisture gave an indication of high fragmentation and spread into near heights. This was supported by low bulk density values for all samples, which shows that the soot can be easily spread by air current to the environment. Presumed volatility level of some of the metals in the soot is an indication that fragmented soot could contribute to environmental pollution by heavy metals. The EDS Spectra shows the presence of impure carbon as the combustion product. This is an indicator of organic pollutant loading. XRF revealed high concentration for Chromium, Copper, Iron, Sulfur, Silicon and Manganese. This study tends to increase awareness of air pollution via domestic emissions within near height environment.

Conflicts of Interest

There is no conflict of interest regarding the publication of this article

Author Contributions

Conceptualization: A.U.I.; S.A.; A.U; Methodology: A.U.I.; R.B.S.; Formal analysis: R.B.S.; Investigation: R.B.S.; Resources: A.U.I, R.B.S., S.A.; A.U.; Data Curation: A.U.I.; Writing – original draft preparation: A.U.I, R.B.S.; A.U.; Writing – review and editing: A.U.I.; S.A.; A.U; Visualization: A.U.I.; S.A.; Project administration: A.U.I.

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