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Comparative Analysis of Heavy Metal Contamination in Regular and Artisanal Petroleum Products in Akwa Ibom State, South-South Nigeria

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Abstract: *The Nigerian government for years has been clamping down on the activities of artisanal crude oil refiners in the Niger Delta. However, the success of such fight is not fully won. Therefore, this study, investigated* the heavy metal content in regularly distributed and artisanal petroleum products in Nigeria, focusing on the concentrations of Lead (Pb), Cadmium (Cd), Chromium (Cr), Nickel (Ni), and Vanadium (V). Samples were analyzed using Atomic Absorption Spectroscopy (AAS), revealing that regularly distributed products contained Pb (0.042 mg/L), Cd (0.003 mg/L), Cr (0.002 mg/L), Ni (0.034 mg/L), and V (0.019 mg/L). In contrast, artisanal petroleum products showed significantly higher *concentrations of Pb (0.212 mg/L), Cd (0.029* mg/L), Cr (0.023 mg/L), Ni (0.157 mg/L), and V (0.137 mg/L). Comparisons with literature values indicated that the levels of Pb and Cd in artisanal products exceeded acceptable limits, raising concerns about their environmental and health impacts. The study highlights the need for stringent quality control measures and the adoption of advanced refining techniques to mitigate the risks associated with the use of artisanal petroleum products in Nigeria.

Keywords*: Diesel, Kerosene, Artisanal, Heavy metal, concentration*

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1.0 Introduction

Petroleum and its fractions contain many metallic compounds in trace amounts depending on the source of the crude and the refinery processes. Trace metallic compositions of crude oil and its fractions have received significant research consideration because it can provide information about the origin and migration of petroleum. Also, some of the metals are catalyst poison, corrosive and can block the process equipment. Vanadium and nickel are the major trace metals in crude oil. Vanadium is a natural component of every crude oil and therefore every fuel oil produced (ISO 14597:1997). Vanadium and its oxides or organometallic are particularly toxic and evaporate during fuel combustions (Corbin *et al*., 2018). The minor metallic compositions in crude oil include copper, cobalt, chromium, manganese, iron, lead, and zinc. Other metals like arsenic, molybdenum, silver, rare-earth elements, and uranium also occur in irregular amounts. Trace elements in hydrocarbon fractions at a certain level can constitute contamination of the hydrocarbon fractions. At elevated temperatures, metal deposits in the engine can cause severe corrosion, deposition on the engine components, and thermal instability resulting in oxidation and deposition of insoluble deposits in the engine (ASTM D7111-16(2021). The presence of trace metals and non-metals in crude oil and petroleum products is destructive, specifically in a refining process. Elevated concentrations of trace metals in hydrocarbon fractions may adversely affect the processing equipment, storage facilities of the petroleum refinery and engine efficiency. These elements are known catalyst poisons and can significantly shorten catalyst life, PIN Petrochemical Chemical and Energy (2024). The traces of heavy metal contents in crude oil are directly associated with the type of crude (light, middle and heavy crude oils) (Barbooti, 2010). The high trace metals content in heavy crude results in high processing costs. Vanadium is an undesired constituent found in crude oil and its products, where it has many side effects in refineries, affects the catalyst activity and causes corrosion problems in plants (Qasim and Sardasht, 2018). Vanadium in crude oil exists mostly as vanadyl ion (VO^{2+}), a form of organometallic complexes and as porphyrins (vanadyl porphyrins). The complexes are formed during the formation of crude oil. Typical problems caused by the presence of vanadium compounds in crude oil are corrosion of the combustion chamber of power plants and changes in catalyst selectivity (Pyrzynska and Wierzbicki 2004).

Trace metals concentrations in hydrocarbon fractions increase on proceeding from light hydrocarbon fractions to heavy petroleum distillates i. e. Gasoline < Kerosene < Diesel < LPFO < Residual fuel). Also, the more viscous the crude oil or petroleum distillates, the more the heavy metal contents. The nature and percentage abundance of heavy metals in crude oil and petroleum products may provide information on the origin, migration, and maturation of raw material of the petroleum products and indication of the regional geochemical prospecting base as well as the processing and storage channels in the refinery (Tijjani *et al.*, 2012). Some trace metals found in crude oil are simply a reflection of those picked up during the migration of the source rock to the reservoir rock. The introduction of drilling mud fluids into the oil well during crude oil extraction can also influence the concentration of metallic impurities in crude oil. The heavy metals in the crude circulate into the petroleum fractions after processing, resulting in the presence of heavy metals in petroleum fractions (Akpoveta and Osakwe, 2014).

Gasoline, kerosene, diesel and other hydrocarbon fractions are in high demand in developing countries due to an increase in population, with a resultant increase in vehicular and industrial activities. This is because refineries e.g. in Nigeria are producing at below-installed capacities or are not functioning at all, which has resulted in the inability to refine enough gasoline to meet local consumption. The gap created by the very low performance of the four Nigerian-owned refineries has resulted in the emergence of artisanal refining activities in the Niger Delta, Nigeria (Udo *et al.,* 2020; Lizabetha Agbakahi, 2022; Yabrade and Tanee, 2016). Although petroleum products produced in the Niger Delta Creek by artisanal refiners have not gone through proper quality control measures to ascertain their agreement with any local or international standards, scientifically they may be efficient in internal combustion engines and mitigates the effect of petroleum product scarcity. Makeshift techniques are used by artisanal refiners in processing crude oil, via thermal cracking, into useful products. These procedures could be unsophisticated and not very safe, however, they could be effective (Udo *et al.,* 2020; Udo *et al.,* 2023a). The petroleum fractions obtained by local refiners are sceptically referred to as "bunkering oil" or adulterated products. Indigenous innovation and ingenuity in harnessing our natural resources should not be outlawed rather it should be regulated, and the products assessed to know if they meet local and international specifications (Udo *et al.,* 2020). Researchers have associated nickel and vanadium emissions from oil combustion with an increased threat in daily mortality. Combustion of residual oil and discharge of used and unused petroleum products into the environment and other petroleum-related activities are sources of elevated heavy metal, metal oxides, polyaromatic hydrocarbons NOx SO_2, CO, and soot into the environment (Ekanem *et al.,* 2021; Udo *et al.,* 2023b; Nwadinigwe *et al.,* 2015; Ephraim-Emmanuel and Enembe, 2023; Oyewale *et al*., 2024; Udo *et al.,* 2018). Also, Nigeria flares more gas per barrel of oil extracted than any other country in the world, contributing to global warming and creating serious health hazards for communities located near gas flares (Pitkin, 2013). The increasing rate of industrial activities, construction activities, transportation and more importantly local and artisanal crude oil refining activities in Niger-Delta Nigeria usually come with damaging effects on the air quality of the region (Odalonu, 2016; Lala *et al.,* 2024).

Analysis of some petroleum products imported into Nigeria recorded high concentrations of heavy metals e.g. lead and copper (Akpoveta and Osakwe 2012). The presence of trace metals and non-metals in crude oil and petroleum products is harmful to the environment if crude oil and petroleum products spill. Contamination of the ecosystem by toxic metals during oil spillage poses serious concern because heavy metals are not biodegradable and are persistent in the ecosystem (Udosen and Awaka-ama, 2005; Nwadinigwe *et al.,* 2014b). Also, among environmental pollutants, heavy metals are of particular concern; due to their potential toxic effect and ability to bioaccumulate in the ecosystems for a long time (Nwadinigwe *et al.,* 2014a). Once metals are introduced and contaminate the environment they will remain for a very long time (Udo *et al.,* 2020). Appraising the concentrations of heavy metals associated with crude oil and petroleum products is necessary for engine malfunction and environmental pollution (Udoetok *et al.,* 2011). The Nigerian government for more than twenty years now has been clamping down on the activities of artisanal crude oil refiners in the Niger Delta. The reasons are because of environmental pollution, oil theft, and the quality of petroleum products produced. But rather chose to import all her petroleum fractions even being the 14th top crude oil producing country in the world and 2nd in Africa. Knowledge of metallic contents of fuel oil circulated in Nigeria can help address the anticipated environmental concern, quality of the refined products and associated health risks. Nevertheless, gasoline, diesel and kerosene produced from artisanal oil refiners still find their way into the petroleum products market. Several studies on artisanal crude oil refined products and their activities have been conducted (Udo *et al.,* 2020; Udo *et al.,* 2023a; *Udo et al.,* 2023b, Iheukwumere *et al*., 2020 and Carpenter, 2023). However, there is still a need for routine periodic quality, and independent checks on petroleum products distributed in the study area to check adulteration, human health risks and environmental pollution. It is on this basis that this research was designed to compare the concentrations of Cr, As, Ni, V, Cd and Fe in Artisanal refined-locally produced diesel and kerosene with Regular distributed (imported) Diesel and Kerosene in Uyo, Niger Delta, Nigeria: a case study of quality.

2.0 Materials and Methods

2.1 Sample collection

Two (2) samples of Artisanal Refined Diesel (ADS) and Artisanal Refined Kerosene were collected at different artisanal refinery sites in Niger Delta, Nigeria. At the Artisanal Refinery sites, the diesel and kerosene samples were added into labelled 2.5 L amber sample bottles through a glass funnel and hermetically sealed. Also, two (2) samples of Regular Distributed Diesel (RDS) and Regular Distributed kerosene (RDS) were collected randomly from two different fuel stations in Uyo Metropolis, Nigeria. The samples were put into airtight amber sample bottles (2.5 L) with a glass stopper. The sample bottles were rinsed with the diesel and kerosene samples to be collected at each fuel station. The sample was introduced into the sample bottle via the dispenser nozzle, labelled and transported to the laboratory for treatment and analysis. The samples were preserved in a refrigerator to avoid volatilization until they were taken for analysis (Udo *et al*., 2023; Udo *et al*., 2020; Udo *et al*., 2023b)

2.2 Sample preparation

10 ml of each sample was digested with a mixture of 20 ml HNO_3 and H_2SO_4 (ratio 4:1) and heated for 4 hours daily in a water bath at a temperature of 80_C for 9 days to ensure complete digestion. The digestion method was adopted to avoid inflammation due to the volatility of the samples (Akpoveta and Osakwe, 2014). The Atomic Absorption Spectrophotometer (AAS) Analyst 400 model was used to demine the concentrations of Cd, Cr, Ni, Ar, Fe and V in the samples (AOAC, 2000).

3.0 Results and Discussion

The concentrations in ppm of Cr, As, Ni, V, Cd and Fe in artisanal refined and regularly distributed diesel and kerosene obtained from Uyo, Niger Delta, Nigeria are shown in Table 1 and in Figs.. 1 and 2.

Table 1: Concentrations of Cr, As, Ni, V, Cd and Fe in Artisanal Refined Kerosene and Diesel with Regular Distributed Diesel and Kerosene

Element	Conc. in Artisanal Diesel (ADS)	Conc. in Regular Diesel (RDS)	Conc. in Artisanal Kerosene (AKS)	Conc. in Regular kerosene (RKS)
Cr	0.166	0.205	0.114	0.014
As	1.421	1.949	0.838	0.000
Ni	0.542	0.644	0.182	0.094
V	1.140	0.000	0.000	0.000
Cd	1.670	1.740	1.145	0.975
Fe	1.514	1.993	0.544	0.117

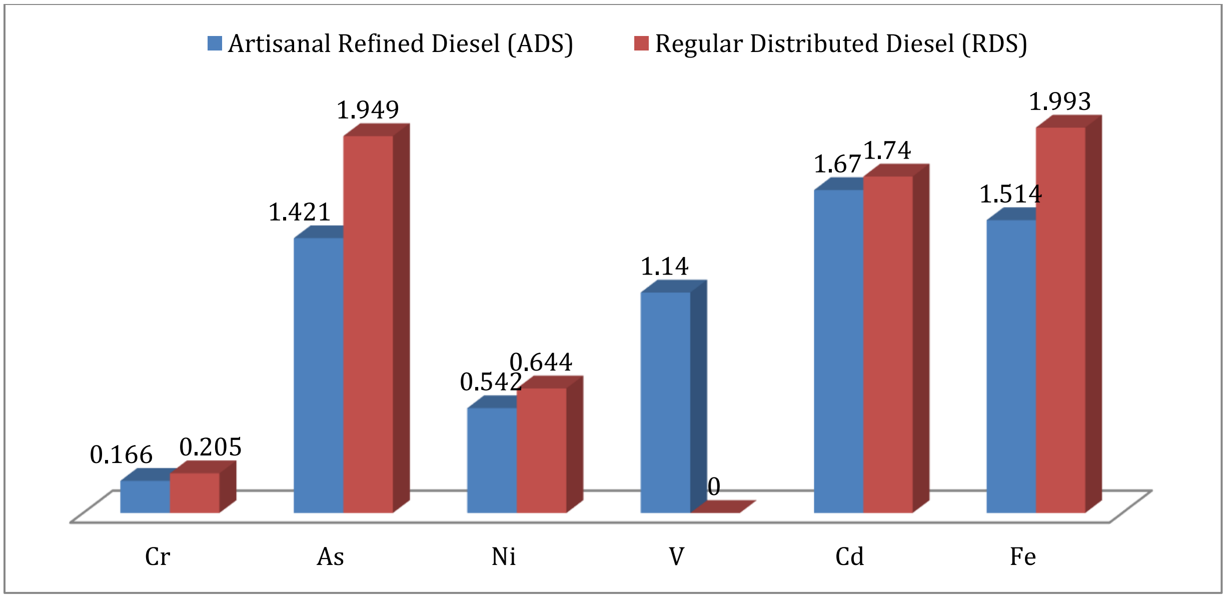

Fig.1 Comparison of the concentrations Cr, As, Ni, V, Cd and Fe in artisanal refined diesel (ADS) and regular distributed diesel (RDS)

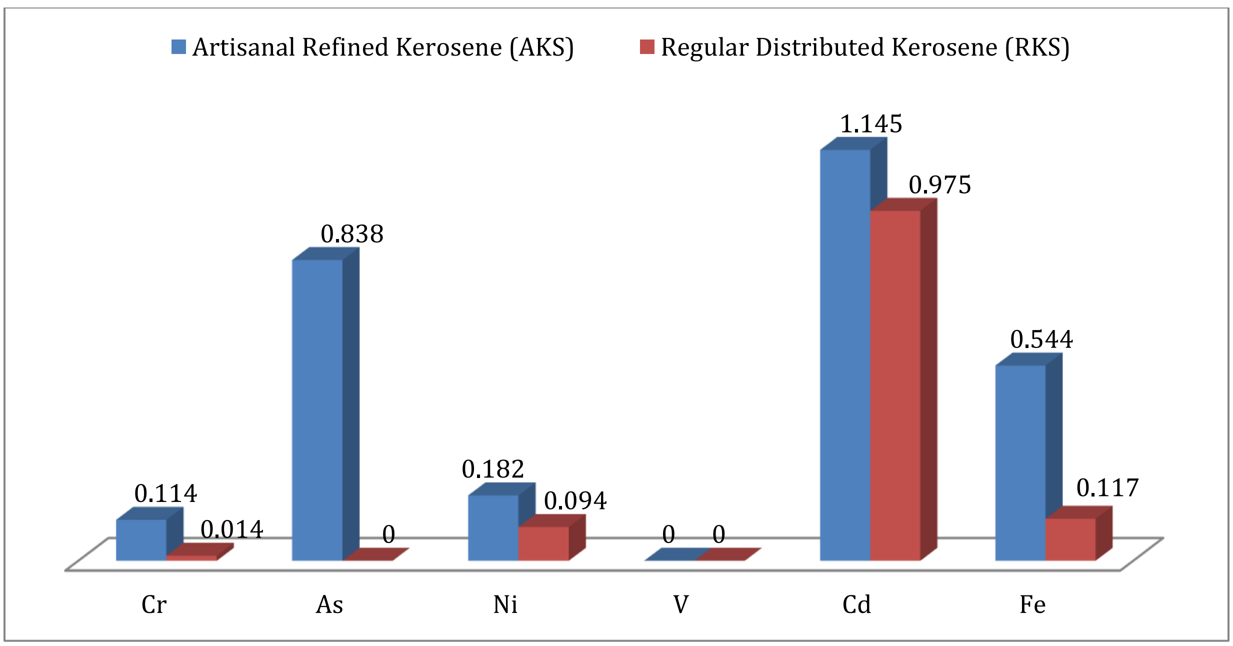

Fig 2: Comparison of the concentrations Cr, As, Ni, V, Cd and Fe in artisanal refined kerosene and regularly distributed kerosene

The concentrations of Cr (0.166), As (1.421), Ni (0.542), V (1.14), Cd (1.67 and Fe(1.514) in artisanal refined (AKS) were higher than the amount of Cr (0.205), As (1.949), Ni (0.644), V(0.00), Cd (1.74) and Fe (1.993) in regular distributed diesel (Figs. 1 and 2). The result indicated slightly elevated concentrations of Cr, As, Ni, V, Cd and Fe in regular distributed diesel (imported diesel) Fig. 1 compared to artisanal refined diesel (locally refined diesel). The enhanced levels of these metals could be attributed to the geochemical location and composition of the original crude, grade of crude oil, source rock, additives, refining process, transport and storage vessels or adulteration (Akpoveta and Osakwe, 2014). Increased concentrations of heavy metals in diesel fuel will result in high heavy metals /metal oxides tailpipe emissions (Pulles *et al.,* 2012). The various amounts of heavy metals present in the investigated diesel samples could be released into the ecosystem during the combustion of the products in internal combustion diesel engines and in case of accidental spillage (Awaka-ama, 2012). These could contribute to an increase in the concentration of heavy metals and their oxides in the ecosystem. The presence of trace metals and nonmetals in crude oil and refined petroleum products is destructive, especially in a refining process (Tijjani *et al.,* 2012). Several toxic metals, including arsenic, cadmium, lead, zinc, antimony, and their compounds, are associated with fine particulate matter in ambient air and are known to be emitted during the combustion of fuel in electric power plants, the engine of vehicles, furnaces and fireplaces (Atiku *et al.,* 2011). Trace amounts of metals such as iron are often found in fuels and can cause issues like equipment corrosion and catalyst deterioration. apart from the gaseous pollutants, diesel combustion also serves as the principal anthropogenic source of trace metals such as Be, Co, Hg, Mo, Ni, Se, Sn, and V. Soot from the exhaust tailpipes of vehicles is one of the main sources of toxic heavy metals into the environment (Nwaedozie and Nyan, 2018; Corbin *et al.,* 2018 and Nomngongo *et al.,* 2014).

The concentrations of Cr(0.114), As (0.838), Ni (0.182), V(0.00), Cd (1.145), Fe (0.544) in artisanal refined kerosene (locally refined kerosene) were higher than the concentrations of Cr(0.014), As (0.00), Ni (0.094), Cd (0.975) and Fe (0.117) in regular distributed kerosene (imported kerosene) except V which was 0.00 in both artisanal and regular distributed kerosene. Tables 1 and Figures. 1-2. Heavy metal concentrations in artisanal refined kerosene and regular kerosene show a wide variation. The disparity may be due to petroleum refining equipment, sample handling procedures and different analytical techniques adopted. The metal contents of Nigerian petroleum products are in general, lower than those of petroleum products from other regions of the world (Tijjani *et al.,* 2012). In a related study Tijjani *et al.*, 2012 reported a value of 1.28 ppm of Fe and 0.18 ppm V, in dual-purpose kerosene DPK. The concentration of heavy metals in kerosene as recorded in this work can cause corrosion of turbines, and distillation towers and when combusted can release toxic oxides of Cd, As and Fe into the environment.

4.0 Conclusion

The study investigated the concentrations of heavy metals (Cr, As, Ni, V, Cd, and Fe) in artisanal refined and regularly distributed diesel and kerosene in Uyo, Niger Delta, Nigeria. Results showed that regular distributed diesel contained slightly higher concentrations of Cr, As, Ni, Cd, and Fe compared to artisanal refined diesel, with values of 0.205 ppm, 1.949 ppm, 0.644 ppm, 1.740 ppm, and 1.993 ppm respectively, while V was only detected in artisanal refined diesel at 1.140 ppm. In the case of kerosene, artisanal refined kerosene exhibited higher concentrations of Cr, As, Ni, Cd, and Fe compared to regular distributed kerosene. These findings were consistent with literature values that indicate the presence of trace metals in petroleum products and highlight the potential environmental and health risks associated with these metals.

The study concluded that both artisanal refined and regularly distributed petroleum products contain significant levels of heavy metals, posing potential environmental and health risks. Regular distributed diesel had slightly higher concentrations of most heavy metals compared to artisanal refined diesel, while artisanal refined kerosene had higher concentrations of metals compared to regular distributed kerosene. These findings are in good agreement with some previous literature, emphasizing the variability in metal content due to factors such as crude source, refining processes, and handling practices.

A consideration of the findings from this study led to the following recommendations

(i) Implementation of enhanced and periodic monitoring and regulation to control the quality of both artisanal and regularly distributed petroleum products,
(ii) Insurance of acceptable safety and environmental standards.
(iii) The adoption of more advanced and environmentally friendly refining techniques among artisanal refiners should be encouraged
(iv) Public awareness campaigns are necessary to educate consumers and refiners about the potential health and environmental risks associated with the use of petroleum products containing high levels of heavy metals.
(v) Directional investment in research and development to explore alternative, cleaner sources of energy and to improve the refining processes for petroleum products is essential.
(vi) Regular environmental assessments should be conducted in areas where petroleum products are refined and distributed

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Data availability

All data used in this study will be readily available to the public

Consent for publication

Not Applicable

Availability of data and materials

The publisher has the right to make the data Public.

Competing interests

The authors declared no conflict of interest.

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Authors' contributions

Udo, Godwin James was involved in conceptualization, Awaka-ama, Joachim Johnson and Nyeneime William Akpanudo, carried out samples collection and laboratorty experiment. Etukudo, Nsikan Jackson. and Udo, Godwin James did the interpretation of results while Uwanta, Emaime Jimmy. and Igwe, Raphael validated the data. Awaka-ama, Joachim Jonhson and Udo, Godwin James did data curation of the manuscript. All authors read and approved the final manuscript.