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Atmospheric Loadings of Lead From Refined Petroleum Products Consumption in Southwestern Nigeria

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Deleterious impact of lead from the use of motor fuels is of major concern. Lead levels of gasoline and diesel samples from States in Southwestern Nigeria were analysed by Atomic Absorption Spectrometer, according to the standard ASTM D3237-97 method. Lead concentrations ranged from 0.491–1.903 mg/L for gasoline and 2.301–10.97 mg/L for diesel. Estimated lead levels in gasoline were below the Department of Petroleum Resources (DPR) Nigeria limit of 3.37 mg/l. Percentage lead contributions from gasoline and diesel fuel consumption were minimal due to various fuel lead-level regulations that have been established over the years.

Keywords: lead, atomic absorption spectrometer, gasoline, diesel

INTRODUCTION

Lead (Pb) is added to fuel as a cost-effective way of increasing the octane number and providing a measure of protection from valve seat wear, which occurs when metal to metal contact of the valve and seat faces cause premature wear (Segui, 2005). The presence of trace metals and nonmetals in the 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, engine of vehicles, furnaces and fire places (Atiku et al., 2011).

Anthropogenic sources of Pb account for its most common sources in the environment largely because it has been widely used throughout history and remains persistent in the environment. These sources also are the most prevalent sources of human Pb exposure, including exposure of the general population and occupational (U.S. Environmental Protection Agency, 1986). A variety of sources and activities contribute to air emissions of Pb, including mobile, area, and stationary sources. In the recent past, the major source of Pb-containing air emissions was the consumption of leaded gasoline in motor vehicles. In 1984 over 89% of an estimated total of 39,000 tons of Pb emitted was attributed to gasoline consumption (U.S. Environmental Protection Agency, 1986). Its global amounts of wet and dry deposition have been estimated to be roughly equal. Of an estimated global atmospheric 410×10^6 kg/year Pb deposition, 208×10^6 kg/year was attributed to wet deposition and 202×10^6

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kg/year to dry deposition (U.S. Environmental Protection Agency, 1986). Their respective annual emissions in Nigeria were 2,790 tons/year (Obioh et al., 1994; Nriagu et al., 1996).

Several methods employed to determine lead levels of refined petroleum products by earlier studies are flame atomic absorption spectrometry (FAAS; Sebor et al., 1982; Osinbanjo et al., 1984; Hammond et al., 1998), electrothermal atomic absorption spectrometry (ET-AAS; Gonzalez et al., 1987; Korn et al., 2007), inductively coupled plasma optical emission spectroscopy (ICP-OES; Borszeki et al., 1992; Brenner et al., 1996; Korn et al., 2007), and inductively coupled plasma mass spectrometry (ICP-MS; Al-Swaidan, 1996; Botto, 2002; Pereira et al., 2010). ICP-MS is highly sensitive multielement technique, but the introduction of organic solvents and the compounds in plasma requires special care similar to ICP-OES, because the organic load may destabilize or extinguish the plasma (Lord, 1991). Other techniques used but not widely reported are aqueous iodine monochloride (Ronald et al., 1967), energy-dispersive X-ray fluorescence spectrometry (Yan et al., 2003), and electroanalytical stripping technique (Munoz et al., 2007).

Over time, as concern over the health effects associated with lead began to grow, health and environmental regulations were enacted to restrict the use of lead in certain products and activities in the world. The new behavioral impairment of children intellectual development is a major concern from lead exposure and can lead to death. The health effects associated with exposure to lead vary widely depending on the level of exposure, the physiological system affected and the receptor (Osuntogun and Koku, 2007). As lead level determination is a primary step for the accurate prediction of lead forming potentials from fuel consumption, the need arises to first determine the present lead level of the refined petroleum products consumed in Southwestern Nigeria to estimate the current contribution of lead air emissions from gasoline and diesel combustion to the national airshed.

EXPERIMENTAL

Study Area

The study area is Southwestern Nigeria, which consists of Lagos, Ogun, Oyo, Osun, Ondo, and Ekiti States (represented as L, G, Y, S, N, and E) for easy identification of samples (Table 1). The area (Figure 1) lies between longitude 2° 31′ and 6° 00′ E and Latitude 6° 21′ and 8° 37′ N. It has a combined land area of 76,352 km^2.

TABLE 1
Sample Coding

State	*Gasoline Sample*	*Diesel Sample*
Osun	S_{g1} S_{g2} S_{g3}	S_{d1} S_{d2} S_{d3}
Oyo	Y_{g1} Y_{g2} Y_{g3}	Y_{d1} Y_{d2} Y_{d3}
Ogun	G_{g1} G_{g2} G_{g3}	G_{d1} G_{d2} G_{d3}
Lagos	L_{g1} L_{g2} L_{g3}	L_{d1} L_{d2} L_{d3}
Ekiti	E_{g1} E_{g2} E_{g3}	E_{d1} E_{d2} E_{d3}
Ondo	N_{g1} N_{g2} N_{g3}	N_{d1} N_{d2} N_{d3}

S = Osun State; Y = Oyo State; G = Ogun State; L = Lagos State; E = Ekiti State; N = Ondo State. Gasoline = g; Diesel = d; Position of filling station = 1, 2, or 3.

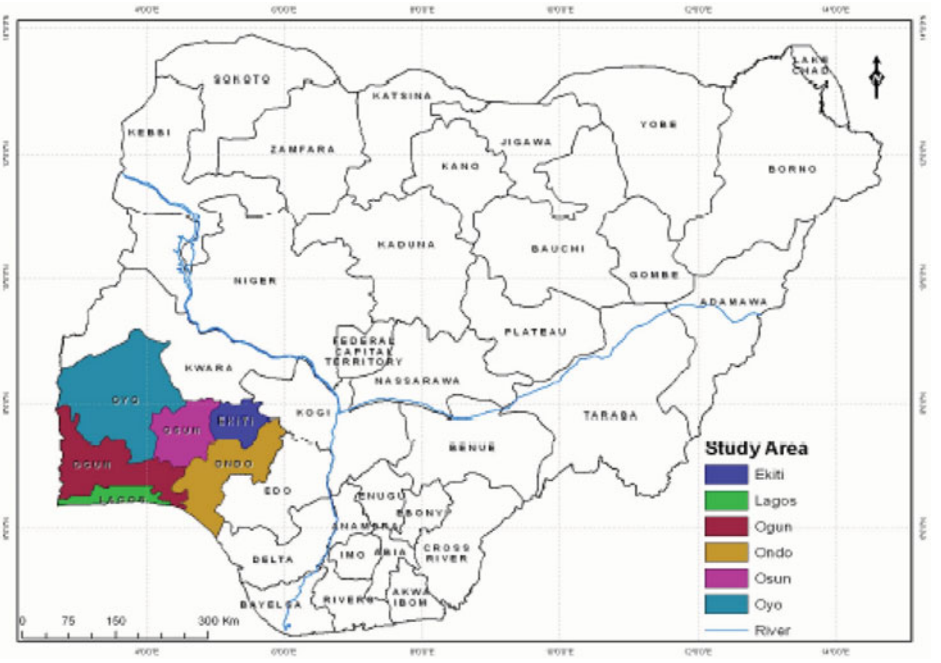

FIGURE 1 The Study Area in Nigeria.

Sample Collection and Storage

Three different gasoline and diesel fuel samples were collected from each of the six states in southwestern Nigeria into plastic bottles and stored in the laboratory at room temperature before analysis. The lead levels of the collected samples were determined using ASTM D3237–97 standard test method for lead in gasoline and diesel by atomic absorption spectrometry (AAS) at wavelength of 283.3 nm. The lead levels of all the petroleum products analyzed in this study were determined on a Model PG-900 atomic absorption spectrometer.

Glassware was deleaded by rinsing with dilute nitric acid (1+1) and then rinsed with distilled water. Glassware was also washed with acetone and dried at $50 \pm 5^{\circ}$C. The average density of all the gasoline samples collected for this study is 0.739 kg/L while the average density of all the diesel samples collected is 0.844 kg/L. In the preparation of the working standards for lead analysis, five different concentrations (0.0, 0.25, 0.50, 0.75, and 2.00 ppm) of lead solution were prepared from the standard stock lead solution (1000 ppm).

In preparing blank for lead in gasoline and diesel analysis, 30 mL of MIBK were added to 10 mL of iso-octane in a 250 mL volumetric flask. A drop of iodine solution was added to the flask and the solution was mixed thoroughly and allowed to react for at 1 min. Also, 5 mL of 1% Aliquat/MIBK solution was added and mixed. The solution in the flask was diluted to 50 mL mark of the flask with MIBK and thoroughly mixed.

Preparation of Gasoline and Diesel Samples for Analysis

In preparing gasoline and diesel samples for analysis, 30 mL of MIBK, 10 mL of gasoline sample, a drop of iodine solution, and 5 mL of iso-octane were added to a 250 mL volumetric flask. The

solution in the flask was mixed thoroughly and reaction was allowed to take place for 3 min before 5 mL of 1% Aliquat/MIBK solution was added.

Standardization and Analysis

The reagent blank was aspirated and the instrument was adjusted to zero. Absorbances of the 0.25, 0.50, 0.75, and 2.00 ppm lead working standards were measured. Gasoline/diesel samples were then aspirated and the absorbance values were recorded. The blank was aspirated between each sample measurement. The instrument automatically prepared a calibration curve by plotting the absorbance of the working standards against their concentrations (ppm) on linear graph. The AAS measured the absorbance of each fuel sample three times and automatically calculated the average absorbance, standard deviation and percentage relative standard deviation. A second order curve equation that relates the concentration of lead in the sample to the absorbance of lead in the sample was also generated by the instrument. The concentration of lead in gasoline and diesel was estimated by manually substituting the absorbance of each fuel sample into the curve equation generated by AAS.

Estimation of Lead Emission

The annual Pb emission from consumption of refined petroleum products was estimated using a combination of annual domestic consumption of refined petroleum products data from Nigerian National Petroleum Corporation annual statistical bulletin (Table 2) and mass balance approach.

TABLE 2
Refined Petroleum Products Consumption in Southwestern Nigeria in Selected Years

Year	*Ekiti*	*Lagos*	*Ogun*	*Ondo*	*Osun*	*Oyo*
Gasoline, L						
1997	2,632,000	952,777,000	189,458,000	74,605,000	57,734,000	181,522,000
1998	8,291,000	1,175,334,000	232,020,000	50,123,000	48,582,000	152,499,000
1999	10,887,000	1,259,751,000	178,944,000	65,348,000	68,573,000	187,742,000
2000	9,342,000	888,348,000	169,110,000	50,242,000	51,309,000	166,312,000
2001	14,915,000	1,491,505,000	302,288,000	91,744,000	93,730,000	277,571,000
2002	25,188,000	1,361,908,000	247,861,000	119,932,000	113,015,000	267,124,000
2005	37,641,000	1,499,452,000	276,292,000	110,478,000	88,248,000	318,263,000
2009	54,356,220	1,449,075,060	286,286,550	106,205,310	124,943,170	340,031,080
2010	32,728,660	635,350,860	140,925,810	77,616,510	100,764,910	183,031,080
2011	32,870,380	486,591,090	82,322,750	72,166,760	52,643,360	150,086,430
Diesel, L						
1997	520,000	324,633,000	40,371,000	22,335,000	9,917,000	66,417,000
1998	1,267,000	300,242,000	32,531,000	12,630,000	5,969,000	44,653,000
1999	1,021,000	382,102,000	40,600,000	12,919,000	10,582,000	49,287,000
2000	755,000	336,428,000	47,663,000	6,960,000	6,960,000	53,418,000
2001	2,835,000	360,493,000	54,286,000	28,098,000	22,327,000	70,009,000
2002	2,835,000	264,609,000	38,076,000	14,644,000	15,509,000	48,813,000
2005	2,081,000	302,351,000	31,959,000	15,110,000	5,518,000	39,805,000
2009	2,208,300	129,987,960	40,612,760	4,263,390	3,550,940	25,646,320
2010	1,373,870	66,185,208	8,438,990	3,375,580	3,939,840	13,875,670
2011	2,980,230	41,397,670	7,379,350	4,587,760	4,794,440	9,284,600

Source: Nigerian National Petroleum Corporation (2011).

Mass balance was the preferred methodology for estimating uncontrolled emissions of Pb from fuel combustion in this study. A worst case scenario proposed by the U.S. Environmental Protection Agency (1986) that suggested that because oil combustion does not generate any bottom ash, it can generally be assumed that 100% of the trace elements present in the fuel are released into the atmosphere upon combustion was adopted in this study. It was generally assumed in this study that the lead content of the fuel was constant for the 10 selected years. The annual lead emissions from gasoline and diesel consumption were calculated using Eq. (1).

$$\begin{aligned} \textit{Annual Emission Rate} &= \textit{Annual Fuel Consumption}\,(L/yr) \\ &\quad * \textit{Lead Content of Fuel}\,(mg/L) \end{aligned} \tag{1}$$

The total annual emission of lead in the Southwestern Nigeria was calculated by summing the annual average lead emissions from all the six Southwestern states of Nigeria.

RESULTS AND DISCUSSION

A standard calibration curve (Figure 2) was generated from standard solution prepared. Statistical analysis showed that the fit gave a good measure of regression with dependency value of 0.9983. The validity of the calibration curve was verified with the closeness of this dependency value to unity. This observation is in consonance with past reports on calibration plot. A second-order Eq. (2) and constants K_0, K_1, and K_2 were generated directly from AAS. This was used to estimate lead concentrations presented in Table 3 by substituting the absorbances of the petroleum product samples.

$$[C] = k_2 * [A]^2 + k_1 * [A] + k_0 \tag{2}$$

Equation factor: $K_2 = 1521.4385$, $K_1 = 80.2213$, $K_0 = -0.0750$, and dependency $= 0.99831$.

As shown in Table 3, the Pb concentrations of the gasoline fuel samples analyzed ranged from 0.491–0.903 mg/L. The maximum observed Pb concentration of 1.903 mg/L was obtained in the L_{g3} gasoline sample from Lagos State while minimum measured Pb concentration of 0.491 mg/L was obtained in the Y_{g1} gasoline sample from Oyo State. However, all the collected gasoline samples from the investigated region analyzed for Pb contained Pb levels below the specified limit of 5 ppm recommended by the DPR.

The results of Pb concentrations in the diesel fuel samples obtained from each of the six states in Southwestern Nigeria are presented in Table 3. The concentrations ranged between 2.30 and

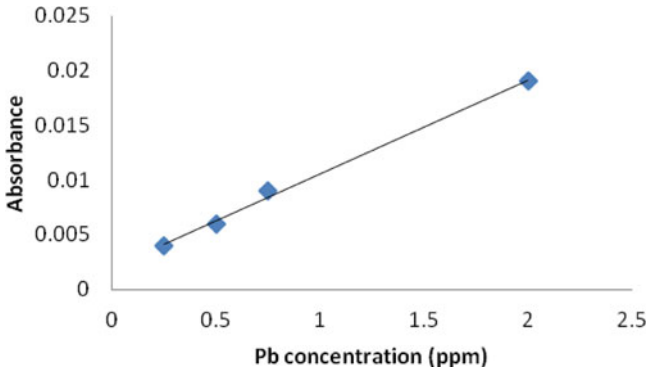

FIGURE 2 Calibration plot for lead in gasoline and diesel fuel.

TABLE 3
Absorbance and Concentration of Lead (Pb) in the Investigated Fuel Samples

Sample	*Average Absorbance*	*Concentration, ppm*	*Concentration, mg/L*	*SD*	*%RSD*
		Gasoline			
S_{g1}	0.015	1.471	1.087	0.0024	2.584
S_{g2}	0.015	1.471	1.087	0.0024	2.593
S_{g3}	0.010	0.879	0.650	0.0023	2.632
Y_{g1}	0.008	0.664	0.491	0.0035	4.041
Y_{g2}	0.015	1.471	1.087	0.0031	3.318
Y_{g3}	0.018	1.862	1.376	0.0004	0.401
G_{g1}	0.013	1.225	0.905	0.0024	2.644
G_{g2}	0.018	1.862	1.376	0.0036	3.745
G_{g3}	0.015	1.471	1.807	0.0024	2.571
L_{g1}	0.021	2.281	1.685	0.0012	1.247
L_{g2}	0.019	1.998	1.477	0.0035	3.508
L_{g3}	0.023	2.575	1.903	0.0054	5.271
E_{g1}	0.022	2.426	1.793	0.0011	1.132
E_{g2}	0.016	1.598	1.181	0.0024	2.560
E_{g3}	0.020	2.138	1.580	0.0022	2.268
N_{g1}	0.020	2.138	1.580	0.0009	0.951
N_{g2}	0.020	2.138	1.580	0.0044	4.487
N_{g3}	0.021	2.281	1.685	0.0012	1.167
		Diesel			
S_{d1}	0.051	7.974	6.730	0.0015	1.322
S_{d2}	0.054	8.693	7.337	0.0007	0.636
S_{d3}	0.039	5.368	4.530	0.0071	6.962
Y_{d1}	0.070	12.99	10.97	0.0017	1.309
Y_{d2}	0.024	2.727	2.301	0.0018	2.019
Y_{d3}	0.043	6.188	5.222	0.0045	4.198
G_{d1}	0.043	6.188	5.222	0.0019	1.783
G_{d2}	0.042	5.978	5.046	0.0026	2.448
G_{d3}	0.061	10.48	8.845	0.0011	0.926
L_{d1}	0.045	6.616	5.584	0.0025	2.279
L_{d2}	0.058	9.696	8.183	0.0043	3.558
L_{d3}	0.037	4.976	4.200	0.0029	2.884
E_{d1}	0.060	10.22	8.622	0.0022	1.774
E_{d2}	0.037	4.976	4.200	0.0036	3.590
E_{d3}	0.038	5.170	4.364	0.0031	3.027
N_{d1}	0.066	11.85	9.999	0.0027	2.083
N_{d2}	0.047	7.056	5.955	0.0017	1.536
N_{d3}	0.051	7.974	6.730	0.0015	1.286

%RSD = percentage relative standard deviation.

10.97 mg/L. The minimum obtained Pb concentration of 2.30 mg/L was obtained in the Y_{d1} diesel sample from Oyo State while the maximum measured Pb concentration of 10.97 mg/L was obtained in the Y_{d2} diesel sample from Oyo State.

The pronounced variations in the Pb concentrations of gasoline and diesel samples could be due to the importations of refined petroleum products of non-uniform quality in Nigeria, storage of petroleum products from various sources over a long period of time, fuel adulteration and use of contaminated tankers. The minimal or negligible deviation of the measured Pb concentrations from standard suggests AAS as an excellent instrumentation method for accurate determination of

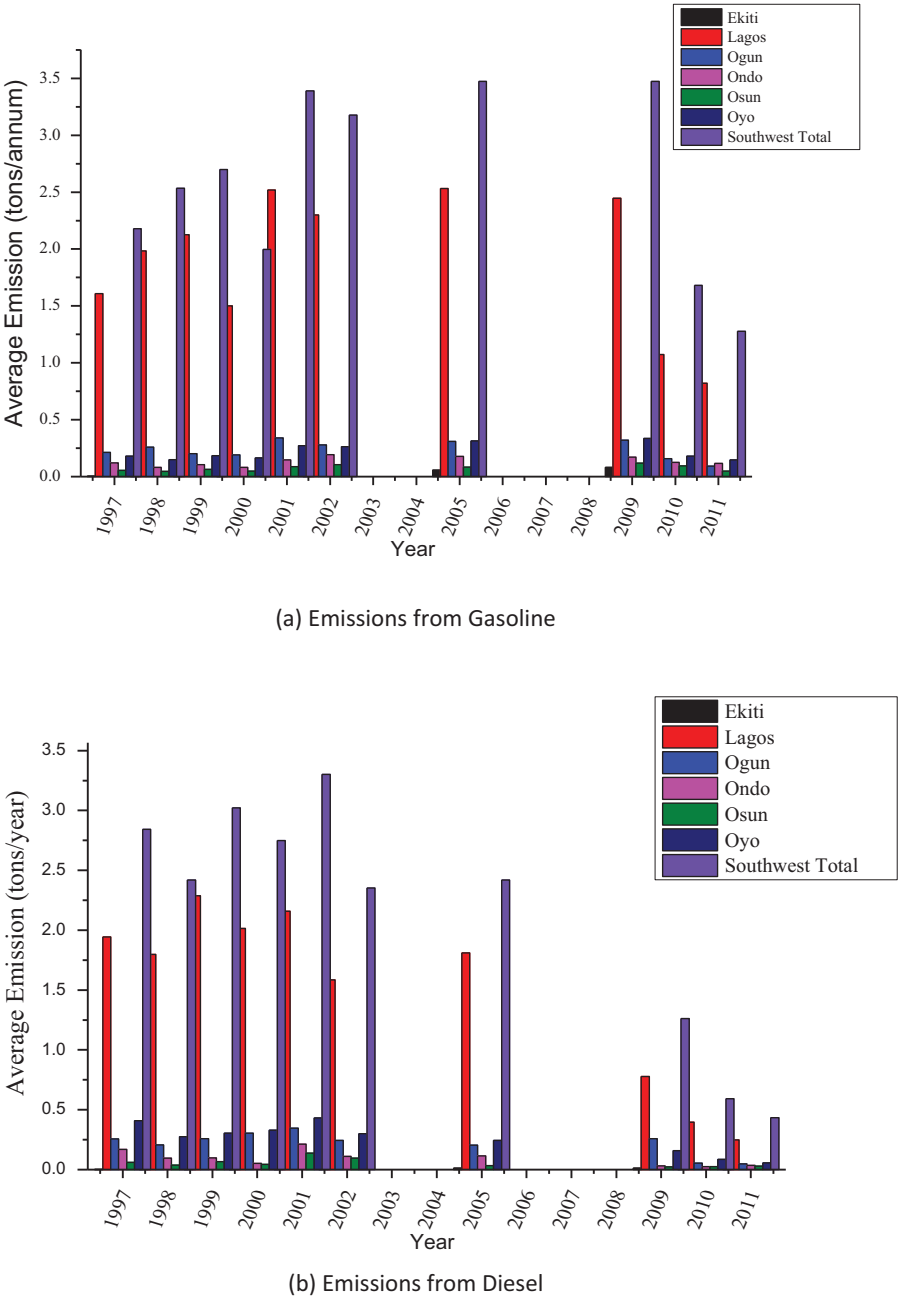

FIGURE 3 Total annual emission of lead from diesel and gasoline consumption in Southwestern Nigeria.

lead in gasoline and diesel fuels. The highest Pb emission of 3.47 tons/year was obtained in 2009, while the lowest Pb emission of 1.28 tons/year was recorded in 2011 (Figure 3). In comparison with the estimated annual total atmospheric lead emissions of 2,790 tons/year in Nigeria reported by Obioh et al. (1994), the result of the percentage contribution of annual Pb emission from gasoline

consumption in Southwestern Nigeria to the annual national Pb emission level shows 0.078% in 1997, 0.091% in 1998, 0.096% in 1999, 0.071% in 2000, 0.121% in 2001, 0.114% in 2002, 0.124% in 2005, 0.124% in 2009, 0.060% in 2010, and 0.046% in 2011.

The result of the annual percentage contribution of Pb emission from diesel consumption in Southwestern Nigeria to the annual national Pb emission level indicates 0.078% in 1997, 0.091% in 1998, 0.096% in 1999, 0.071% in 2000, 0.121% in 2001, 0.114% in 2002, 0.124% in 2005, 0.124% in 2009, 0.060% in 2010, and 0.046% in 2011 using the estimated annual total atmospheric lead emissions of 2,790 tons/year in Nigeria reported by Obioh et al (1994) as the standard national Pb emission level. Figure 3 shows that the lowest Pb emission of 0.434 ton/year was recorded in 2011 while the highest Pb emission of 3.302 tons/year was obtained in 2001. The percentage contribution of lead emission from fuel consumption estimated in this study to national lead emissions level is very low because of the various fuel lead level regulations that have been established over the years to phase out the consumption of leaded fuel. It is therefore expected that the current national lead emissions level would have dropped drastically compared to the levels reported by Obioh et al. (1994) and Nriagu et al. (1996).

CONCLUSIONS

In this study, atmospheric loadings of lead from gasoline and diesel consumption in Southwestern Nigeria for 10 selected years were investigated. Laboratory experiments were carried out to determine the lead levels of in gasoline and diesel consumed in the part of the country. The petroleum product samples analyzed contained a wide variety of lead concentrations ranging from 0.491–1.903 mg/L for gasoline and 2.301–10.97 mg/L for diesel. The obtained results showed total level of compliance of the lead level in all the gasoline samples from Southwestern Nigeria with the lead limit of 3.37 mg/L (5 ppm) recommended by the Department of Petroleum Resources (DPR) Nigeria. However, no lead limit was specified by DPR for the lead level of diesel fuel.

The percentage contributions of lead emissions to the national annual lead emissions level were found to be very minimal. The study suggested that to mitigate the air emissions of lead, a cleaner technology control approach should be adopted in the form of fuel modification. As lead emission is proportional to the lead contents of the fuel, the use of unleaded fuel must be enforced. It also, suggested that future work should be extended to investigate the contribution of lead emissions from consumption of refined petroleum products in other zones and Federal Capital Territory of Nigeria to the national emission levels. The results of this study will further help to identify appropriate control technology for the mitigation of air emissions of lead from refined petroleum products in the country.

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