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Atmospheric particulate fractions from Nigerian crude oil spillage

Adewole Johnson Adesanmi[a], Oyetunji Babatunde Okedere[b,*], Jacob Ademola Sonibare[c], Francis Boluwaji Elehinafe[d], Bamidele Sunday Fakinle[e]

[a] Laboratory Research & Bioresources Development Centre, Akoko, Nigeria
[b] Department of Chemical Engineering, Osun State University, Osogbo, Nigeria
[c] Environmental Engineering Laboratory, Department of Chemical Engineering, Obafemi Awolowo University, Ile-Ife, Nigeria
[d] Department of Chemical Engineering, Covenant University, Ota, Nigeria
[e] Department of Chemical Engineering, Landmark University, Omuaran, Nigeria

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ABSTRACT

Laboratory simulations of the spill behaviors of three different Nigerian crude oil samples over three media (fresh water, sea water and soil) were carried out with a view to determining the effect of crude oil spillage on emission of fine ($PM_{2.5}$) and inhalable particulates fractions (PM_{10}). The spillage experiments were carried out in an environmental test box fitted with equipment to regulate the micro climatic conditions (temperature and Relative humidity). The maximum concentrations of $PM_{2.5}$ were 711, 689 and 680 μgm^{-3} while those of PM_{10} were 972, 946 and 940 μgm^{-3} for sample A, B and C respectively. These peak concentrations were obtained for spill conditions corresponding to fresh water at 45 °C and 49% relative humidity. The least concentrations of $PM_{2.5}$ and PM_{10} emitted across all samples were 63 μgm^{-3} and 256 μgm^{-3} respectively and these corresponded to spillage over sea water at 15 °C and 80% relative humidity. These concentrations clearly exceeded the short time averaging period (24 h) standards set for $PM_{2.5}$ and PM_{10} by the United States Environmental Protection Agency. Incessant crude oil spillages in the Nigerian oil fields are therefore predicted to cause degradation of air quality within a short duration from the spill.

1. Introduction

Atmospheric particulates (PM) are of major air pollution concern globally because of the hazards they pose to public health (Burki, 2019; Kumar et al., 2021). By intrinsic properties, they can cause damage to the human health or aggravate disease conditions most especially in vulnerable groups such are infants, aged or people with underlying respiratory diseases. Apart from their hazardous intrinsic properties, they can also serve as conveyance media by which other harmful pollutants enter the human body. This is possible when such pollutants are adsorbed unto the surface of the particles. Several reports on the harmful effects of atmospheric particulates abound in the literature (Kumar et al., 2021; Dockery and Pope, 1994; Bell et al., 2014). Cardiovascular and respiratory functions of the body are the most affected by inhaled air-borne particles (Kumar et al., 2021; Kappos et al., 2004; Heal et al., 2012; Guo et al., 2019; Yang et al., 2019; Yang et al., 2020).

The particles fractions are usually categorized as ultra fine, fine, coarse or inhalable depending on the aerodynamic diameter which is commonly expressed in micro meter (μm). According to Moreno-Rios et al. (2021) and Kan et al. (2007), ultra fine ($PM_{0.1}$) are the fraction of particles with aerodynamic diameter (D_A) of less than 0.1 μm; fine fraction of particles ($PM_{2.5}$) have D_A of less than 2.5 μm; coarse fractions ($PM_{10-2.5}$) have D_A between 2.5 and 10 μm while inhalable fractions (PM_{10}) have DA of less than 10 μm. The extent to which atmospheric particulates affect human health depends on the diameter of the particle, the nature of their surfaces, composition, the region where they are deposited and the extent of vulnerability of the people that are exposed to their emissions. Generally, ultra fine and fine particles are the range of particle sizes that have been predominantly linked to negative public health impacts. While PM_{10} is majorly deposited in the respiratory tracts, ultra fine particles get deeper into the lungs and the rest of the human system (Moreno-Rios et al., 2021). There are toxicological and epidemiological evidences to the cytotoxic, inflammatory, cardiovascular and respiratory effects of PM (Jalava et al., 2015; Hu et al., 2018).

The sources contributing to atmospheric PM are ubiquitous and they range from natural to anthropogenic. Most anthropogenic sources of PM are combustion related and they include emissions from transport systems (automobiles, airplanes and ships) (Fridell et al., 2008; Fakinle et al., 2013), industrial processes and utility stacks (Okedere et al., 2015; Okedere et al., 2018; Oki et al., 2018; Okedere et al., 2021), forest and biomass burning (Okedere et al., 2017), waste incineration

* Corresponding author.
E-mail address: oyetunji.okedere@uniosun.edu.ng (O.B. Okedere).

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(Adesina et al., 2018; Okedere et al., 2019; Adesina et al., 2020; Fakinle et al., 2020), petroleum refining (Oladimeji et al., 2015a; Oladimeji et al., 2015b), gas flaring (Akeredolu, 1989), thermal plants (Sonibare, 2010), domestic cooking (Fakinle et al., 2020) and so on. Several studies have been undertaken on the contribution of these combustion sources to atmospheric levels of particulate in Nigeria but little is known on the particulate emission characteristics of Nigerian crude oil during spillage.

Crude oil spillage is another potential source of particulate emission to the environment and crude oil being the predominant source of income for Nigerian government, excessive pressure on exploration, drilling, processing and transportation exist which always result in spillages. Mohamadi et al. (2016), indicated that the Niger Delta region of Nigeria is facing serious environmental challenge due to frequent oil spills. Up to 163 incidents of crude oil spillage were reported for River State within a space of one calendar year (2011–2012) (Mohamadi et al., 2016). According to a much recent report by Akinwumiju et al. (2020), close to 70% of the approximately 8000 cases of crude oil spillage in Nigeria between 2006 and 2019 occurred onshore. These spillages are frequently experienced in the Niger Delta region of the country where the crude deposit and other oil installations are located (Mohamadi et al., 2016).

Crude oil spillage could arise from faulty equipment, accidents involving vessels, and barges, damages to pipelines and human error (Arigo, 2013; Mawuli et al., 2016). Properties of crude oil such as specific gravity (SG), American Petroleum Institute (API) gravity and viscosity influence their flow behaviours, volatility, evaporation rates and consequently their tendencies to emit air pollutants when crude oil spillage occurs (Udeme and Etim, 2012). Crude oils may be classified on the basis of their API gravity as very heavy (API $<$ 10), heavy (10 $<$ API $<$ 30), medium (30 $<$ API $<$ 40) and light (API $>$ 40) (Al-Dahhan and Mahmood, 2019). Despite the huge volume of crude oil spillage in Nigeria, there is dearth of information on their contribution to atmospheric level of particulates and consequently public health impacts of these emissions. There is no doubt that several studies have implicated crude oil spills on soil degradation and water contamination. The map of Nigeria showing the different regions and a typical scenario of environmental degradation by crude oil spill in South-South (Niger Delta) region of Nigeria are shown in Fig. 1 and Fig. 2 respectively (Akinlua et al., 2015; Kadafa, 2012). The present study examines the emission of fine and inhalable particulates ($PM_{2.5}$ and PM_{10}) emitted during laboratory simulated crude oil spillage experiments with a view to determining if such spillages would emit particulates sufficient enough to pose human or environmental health hazards.

2. Materials and method

2.1. Crude oil samples and analyses

Three different crude oil samples labeled A, B, and C were obtained from Niger Delta Petroleum Resources (NDPR) and SEPLAT which are major oil industry players in Nigeria. According to information from NDPR and SEPLAT, the oil samples were from different oil wells located in Oghele (Rivers State), Bayelsa States and Sapele (Delta State) of Nigeria respectively. Letters of authority from Nigeria National Petroleum Commission and Department of Petroleum Resources were taken to these companies to collect crude oil samples. The samples were analyzed for density, specific gravity, API gravity and viscosity. The density (ρ), specific gravity (SG) and API gravity were determined with Eqs. (1), (2) and (3) (Al-Dahhan and Mahmood, 2019; Daubert and Danner, 1997) while the viscosity was measured with ISO 9001 compliant Cannon-Fenske reverse flow viscometer according to ASTM D444 and ASTM D52515 standard methods (Fan, 2001; Fitch, 2013).

$$\rho = \frac{\text{Mass of Crude Oil}}{Volume\ of\ cude\ Oil} \quad (1)$$

Table 1
Simulated environmental chamber climatic conditions.

	*T1	*T2	*T3	*T4
Temperature (°C)	15	25	35	45
Relative Humidity (%)	80	71	68	49

* T1-T4 (Test conditions).

$$SG = \frac{Density\ of\ Crude\ Oil}{Density\ of\ Water} \quad (2)$$

$$\text{API gravity} = \frac{141.5}{SG} - 131.5 \quad (3)$$

2.2. Soil and water analysis

The soil samples used for the experiments were obtained at a depth of 0–15 cm from an oil producing community in Bayelsa State, Nigeria as recommended by Song et al. (1990). The fresh water and sea water samples were taken from Pattani River in Bayelsa State and open sea in Lagos State respectively. The samples were taken in these areas to obtain conditions that are close to real life spillage since crude spill can take place either onshore or offshore. Soil properties including texture, bulk density, particle density, organic matter content and total porosity were determined for the soil following procedures described by Black et al. (1979), Juo (1978) and Ayotamuno and Kogbara (2007). The pH of the water sample was also noted.

2.3. Description of environmental chamber and experimental procedure

Particulate emission characterizations were carried out in an air tight environmental test box of dimension 0.6 x 0.6 x 1 m equipped with a 0.25 horse power cooling unit, speed fan of 1000 rpm and 1000 W heat generating unit for regulation of climatic conditions within the chamber. Measurement of climatic conditions before and during each experimental run was achieved with Kestrel 4500 weather tracker. The device has a resolution of 0.1° and 0.1% and an accuracy of ± 1 °C and ± 2% for temperature and relative humidity respectively (Richard Paul Russell Limited (RPR), 2008). The chamber was designed to accommodate stainless steel container capable of holding 6.0 liters of crude oil at its base. Three of such stainless steel containers were acquired to run spill experiments on crude oil samples A, B and C so labeled based on their API gravity. The test box has a tempered glass on one of its sides to aid visual inspection and recording of data. The wind supply device was regulated and maintained at 2.4 m/s.

To simulate the emission of $PM_{2.5}$ and PM_{10} during crude oil spillage, 593 ml of each of crude oil samples (A, B and C) were spilled over three different media (fresh water, sea water and soil) created inside the steel container in the test box and independently investigated under the same climatic conditions as summarized in Table 1. The climatic conditions were chosen because they are good representations of average temperature, humidity and wind speeds in Nigeria (Eludoyin et al., 2013). The steel container in the environmental chamber was first filled with 4 liters of fresh water followed by gradual pouring (spilling) of 593 ml of sample over the surface of the water. Subsequent experiments involved replacement of the spill medium (fresh water) with 4 liters of seawater and later 4 kg of soil. AEROCET 531S mass monitor/particle counter (Item number MO-AEROCET-531S) from Met-One instruments was used to measure the particle concentrations. The equipment has concentration range of 0 – 1000 μgm^{-3}, resolution of 0.1 $\mu g/m^3$, a time sampling of 5 min, and flow rate of 2.83 l/min (Met-One Instruments 2014). Particulate matter $PM_{2.5}$ and PM_{10} representing particles of diameter less than 2.5 μm and 10 μm respectively were considered for investigation due to their links with morbidity of individuals. To measure $PM_{2.5}$ and PM_{10} during spillage, the monitor was placed at 20 cm from the surface of the

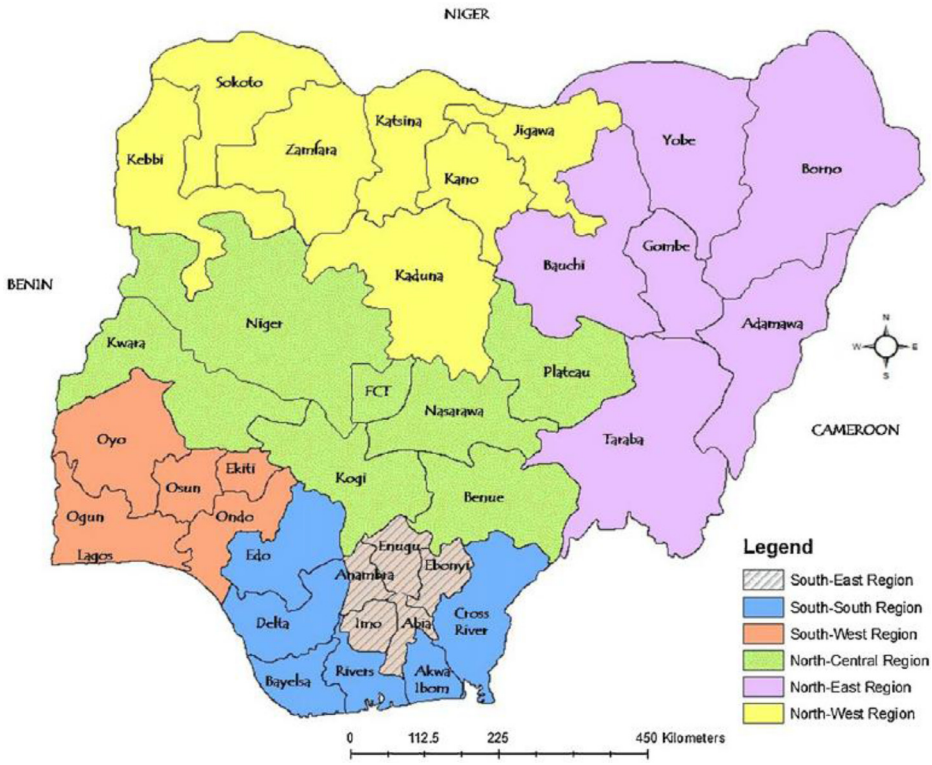

Fig. 1. Map of Nigeria showing the States and Regions with South-South in blue (Akinlua et al., 2015).

Fig. 2. Typical scene of environmental degradation by oil spillage in south-south region of Nigeria (Kadafa, 2012).

spillage in the test box, switched on while the measured concentrations were read directly from the screen after particle capturing.

3. Results and discussion

3.1. Properties of crude oil and the spill media

The physical properties of the three crude oil samples investigated are presented in Table 3. The specific gravity, API gravity and viscosity of the samples ranged between 0.83–0.86, 32.00 -38.00 and 7.17 -7.41 mm^2/s respectively and are thus classified as light crude oils. The large API gravity implied that the crude oils tend towards light crude classifications (Al-Dahhan and Mahmood, 2019). In addition, the relatively lower viscosities are indications of high volatility and evaporation (Udeme and Etim, 2012). These properties will aid rapid flow and evaporation of crude oil in the environment during spillage thereby increasing environmental pollution tendencies. The properties of the soil sample used as spill medium are as summarized in Table 2. The mean bulk density, porosity, particle density and organic matter contents were 1.25 g/cm^3, 51.1%, 2.68 g/cm^3 and 2.48% respectively indicating a relatively high resistance to liquid absorption, a property which may aid flow and spread of spilled the oil.

3.2. Concentration of $PM_{2.5}$ and PM_{10} particulates from crude oil spills

The measured concentrations of $PM_{2.5}$ and PM_{10} from the simulated spillage experiments involving sample A over sea water, fresh water and soil under different climatic conditions are as represented in Tables 4 and 5. The mass concentrations of the aerosol emitted from spillage of sample A at temperature 15 °C and relative humidity of 80% were in the range of 84 – 103 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 307 – 341 $\frac{\mu g}{m^3}$ for PM_{10} while at temperature 25 °C and relative humidity of 71% they were in the range of 78 – 81 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 271 – 345 $\frac{\mu g}{m^3}$ for PM_{10} respectively. In the same vein, the observed mass concentrations of aerosols from sample A at temperature 35 °C and relative humidity of 65% were in the range of 160 – 180 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 328 – 380 $\frac{\mu g}{m^3}$ for PM_{10} while they were in the range of 100 – 710 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 381 – 970 $\frac{\mu g}{m^3}$ for PM_{10} at temperature 45 °C and relative humidity of 49%.

In another experiment involving spillage of sample B, the concentrations of the emitted aerosols are also depicted in Tables 4 and 5. At 15 °C and relative humidity of 80%, the mass concentrations were in the range of 78 – 101 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 301 – 361 $\frac{\mu g}{m^3}$ for PM_{10} while they were in the range of 71 – 78 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 241 – 335 $\frac{\mu g}{m^3}$ for PM_{10} at 25 °C and relative humidity of 71%. At 35 °C and relative humidity of 68%, the emitted particulates were in the range of 133 – 161 $\frac{\mu g}{m^3}$ for $PM_{2.5,}$ and 309 – 357 $\frac{\mu g}{m^3}$ for PM_{10} while at 45 °C and relative humidity of 49% they were in the range of 69 – 516 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 362 – 965 $\frac{\mu g}{m^3}$ for $PM_{10.}$

The spill behaviors of sample C in terms of aerosol emission are presented in Tables 4 and 5. The mass concentration of the aerosol emitted at 15 °C and relative humidity of 80% were in the range of 72 – 95 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 296 – 342 $\frac{\mu g}{m^3}$ for PM_{10} while at 25 °C and relative humidity of 71% they were in the range of 62 – 71 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and

Table 2
Soil and water properties.

Bulk Density (g/cm3)	Porosity (%)	Particle Density (g/cm3)	Organic Matter Content(%)	pH of Fresh Water	pH of Sea Water
1.25 ± 0.02	51.10 ± 3.50	2.68 ± 0.10	2.48 ± 0.03	6.90	7.80

Table 3
Properties of crude oil samples.

Crude oil samples	Density g/ml	Specific gravity	Viscosity (mm^2/s)	API gravity
A	0.85 ± 0.06	0.83	7.17 ± 1.12	38
B	0.87 ± 0.04	0.84	7.23 ± 1.50	34
C	0.88 ± 0.02	0.86	7.41 ± 1.21	32

Table 4
Concentrations of $PM_{2.5}$ from crude oil spill over different spill media ($\mu g/m^3$).

		Sample A			Sample B			Sample C	
Temperature /Relative Humidity	Sea Water	Fresh Water	Soil	Sea Water	Fresh Water	Soil	Sea Water	Fresh Water	Soil
15 °C/80%	78.4 ± 5.6	78.8 ± 4.2	81.0 ± 7.3	72.4 ± 6.8	72.8 ± 3.1	75.0 ± 2.8	63.4 ± 6.1	63.8 ± 2.1	66.0 ± 5.6
25 °C/71%	103.3 ± 7.1	103.6 ± 6.3	85.0 ± 9.2	97.3 ± 7.3	97.6 ± 5.2	79.0 ± 4.1	88.3 ± 5.8	88.6 ± 7.4	70.0 ± 6.2
35 °C/68%	160.1 ± 6.8	160.5 ± 7.1	180.7 ± 7.1	138.3 ± 8.1	138.5 ± 6.1	158.7 ± 5.2	129.1 ± 4.9	129.5 ± 8.2	149.7 ± 6.8
45 °C/49%	710.6 ± 9.2	710.9 ± 6.2	100.0 ± 3.4	688.6 ± 10.2	688.9 ± 8.4	511.3 ± 9.2	679.6±.8.1	679.9 ± 7.9	69.0 ± 3.7

Table 5
Concentrations of PM_{10} from crude oil spill over different spill media ($\mu g/m^3$).

		Sample A			Sample B			Sample C	
Temperature/ Relative Humidity	Sea Water	Fresh Water	Soil	Sea Water	Fresh Water	Soil	Sea Water	Fresh Water	Soil
15 °C/80%	271.3 ± 9.4	271.7 ± 11.3	345.4 ± 8.3	265.3 ± 8.7	265.7 ± 10.0	339.4 ± 12.4	256.3 ± 8.9	256.7 ± 11.2	330.4 ± 12.1
25 °C/71%	307.5 ± 8.6	307.9 ± 8.8	341.0 ± 7.9	301.5 ± 10.3	301.9 ± 11.5	335.0 ± 12.1	292.5 ± 9.3	292.9 ± 7.9	326.0 ± 9.8
35 °C/68%	328.0 ± 7.9	328.4 ± 10.2	480.2 ± 10.1	306.0 ± 9.6	306.4 ± 9.8	458.2 ± 10.1	297.0 ± 7.8	297.4 ± 10.1	449.2 ± 10.4
45 °C/49%	961.2 ± 11.2	971.6 ± 12.5	391.0 ± 9.4	939.2 ± 11.9	949.6 ± 12.7	369.4 ± 9.1	930.2 ± 13.5	940.6 ± 11.7	360.4 ± 6.8

235 – 313 $\frac{\mu g}{m^3}$ for PM_{10}. At a much higher temperature and relative humidity of 35 °C and 65% respectively, the mass concentrations of the emitted aerosols were in the range of 121 – 150 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 301 – 334 $\frac{\mu g}{m^3}$ for PM_{10} while at 45 °C and relative humidity of 49%, they ranges were 61 – 501 $\frac{\mu g}{m^3}$ for $PM_{2.5}$ and 343 – 928 $\frac{\mu g}{m^3}$ for PM_{10}.

For a uniform spill volume of crude oil of 593 cm^3 over three different spill media in the experimental chamber, the highest mass concentrations of $PM_{2.5}$ and PM_{10} emitted were recorded from crude oil sample A while samples B and C also follow in that order. Results showed that the emission of particulate from crude oil spillage is largely dependent on the physical properties of the crude oil and the prevailing atmospheric condition. For instance, sample A with the least viscosity and specific gravity emitted the most $PM_{2.5}$ and PM_{10} which progressively increased as the temperature and humidity were varied from 15 °C, 80% to 45 °C, 49% respectively. This is expected because viscosity as a fluid property decreases with increase in temperature. The increase in temperature from 15 °C to 45 °C generally reduced the viscosities of all the crude oil samples, hence, increase in rate of evaporation and consequently the emission of aerosols (Udeme and Etim, 2012).

Among these PM categories, there have been several documentations about the health risks associated with $PM_{2.5}$ and PM_{10}. In the absence of national standard for $PM_{2.5}$ and PM_{10} in Nigeria, the United States national standard for ambient air quality for a very short (24 h) averaging period concentrations of $PM_{2.5}$ and PM_{10} which are 35 μgm^{-3} and 150 μgm^{-3} respectively were used as the basis for comparison (United States Environmental Protection Agency (USEPA) 2019). The aerosol concentrations obtained in this study clearly exceeded the short time averaging period (24 h) standards set for $PM_{2.5}$ and PM_{10} in the United States of America. Incessant crude oil spillages in the Nigerian oil fields are therefore predicted to cause degradation of air quality within a short duration from the spill.

The allowable ambient air concentrations of particulate matter emanating from crude oil spills may even be lower than the United States standards for ordinary $PM_{2.5}$ and PM_{10} because such particulates are organic aerosols and usually contain hazardous compounds such as benzene, toluene, ethyl-benzene, xylene and polycyclic aromatic hydrocarbons (PAHs). These hazardous organic pollutants have been reported by United States Agency for Toxic Substances and Disease Registry (ATSDR) as having carcinogenic tendencies. Hence, in addition to the intrinsic health impairment risks associated with $PM_{2.5}$ and PM_{10}, the PMs investigated in this study may also serve as vehicles for conveyance of carcinogenic pollutants into the human body (Lim et al., 2021). Such hazardous aerosols will raise the ambient concentrations of the associated organic pollutants with further tendencies to negatively impact public health most especially among the vulnerable groups. The recommended limits for organic aerosols in ambient air are usually lower than those of ordinary PM. According to Ana et al. (2012), the recommended standard for PAHs in ambient air is as low as 12 ng/m^3. The emitted range of concentrations of $PM_{2.5}$ and PM_{10} obtained in this study are therefore considered too large because of the possibility of their impregnation with hazardous organic pollutants which have carcinogenic, mutagenic and teratogenic properties.

4. Conclusion

In this study, crude oil spillage experiments have been simulated in an environmental chamber. Results from spillage of all the crude oil samples of Nigerian origin showed that atmospheric loading of aerosols from the crude oil spillage could be large enough to impact public health. The obtained concentrations of particulates exceeded the 24 h averaging period concentration set for the pollutants in the United States. The situation is even more worrisome because aerosols

from crude oil usually contain hazardous organic compounds which have been shown to possess carcinogenic and mutagenic properties. Given these results, it is recommended that appropriate steps be taken to minimize frequency of crude oil spillage and when oil spill occurs, the response time for cleanup should be improved upon.

Declaration of Competing Interest

There authors affirm that there is no conflict of interest.

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