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Ambient Air Emission Profiles of Polycyclic Aromatic Hydrocarbons around a Typical University Power House in Nigeria

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ABSTRACT

The level of electricity generation in Nigeria is presently grossly inadequate. Consequently, the productive sectors of the economy do not presently enjoy an uninterrupted supply of electrical energy from the country's national grid and the downtime is in most cases too much. Using diesel-fueled electric power generators has become an attractive alternative to meet the shortfall in the energy supply gap. In this study, the levels of polycyclic aromatic hydrocarbons in the vicinity of the diesel-fueled electric power generators operated by Osun State University, Osogbo, Nigeria were investigated with a view to ascertain the priority levels and carcinogenic PAHs. Since the generators were recently relocated, the study covered the old and the new sites. Polyurethane foam passive samplers were deployed at the old and new sites for 28 days. They were subjected to Soxhlet extraction using dichloromethane to extract the PAHs from the PUFs. Cleaning up and elution were done with 5 g of silica gel column and 40 mL 1:1, DCM:Hexane followed by concentration in a rotary evaporator using a stream of nitrogen. Gas chromatography-mass spectrometry (GC-MS) analysis was used to determine of PAHs. The Σ priority PAHs around the old and new sites were in the range of 4.43-5.44 $\mu\text{g}/\text{m}^3$ and 4.47-5.35 $\mu\text{g}/\text{m}^3$, respectively. The Σ carcinogenic PAHs at the sites were 2.98-3.42 $\mu\text{g}/\text{m}^3$ and 3.35 - 4.17 $\mu\text{g}/\text{m}^3$. Higher molecular weight PAHs dominated the measured PAHs with benzo[a]pyrene being the most significant contributor to the computed total toxicity equivalence (TTEQ). The PAHs diagnostic ratios affirmed that the measured PAHs were from diesel-related sources. The levels of the PAHs obtained in this study are above permissible limits specified for ambient air; hence, the air around the power plant is considered unsafe.

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INTRODUCTION

Energy availability in the form of electrical power is crucial for human society's development and economic activity. Small, medium and large-scale enterprises need regular supply of power to remain in business. Research centers among those universities also need stable supply of power to conduct meaningful laboratory studies. In Nigeria, a reliable supply of electrical energy is still a severe challenge due to insufficient energy generation. The country presently has nearly 200 million people and generates only about 4000 MW of electrical energy; hence, regular supply and availability are not guaranteed (Akhator et al., 2019 and Ajewole et al., 2021). This has led to heavy reliance on backup gasoline or diesel generators of varying sizes and capacities by almost all segments of society depending on their need and financial capacity (Oseni, 2016; Adeniran et al., 2017 and Okedere et al., 2021). A manufacturing firm can only operate successfully in Nigeria with a backup generator. A very glaring example is the deployment of backup diesel generators at the several thousand base transceiver stations of mobile telecommunication industries in Nigeria (Okedere et al., 2018 Okedere and Oyelami, 2021).

Apart from economic considerations regarding the cost of diesel, environmental sustainability associated with burning fossil fuels for energy generation is a crucial factor that has been alluded to in previous studies (Fakinle et al., 2013; Okedere et al., 2015; Okedere and Elehinafe, 2016; Fakinle et al., 2018a). Energy production systems such as diesel-fueled internal combustion engines contribute significantly to the emission of pollutants to the environment (Fakinle et al. 2018b; Lala et al., 2019).

Among several environmental contaminants, there appears to be a renewed interest in polycyclic aromatic hydrocarbons (PAHs). The reason for this is not far-fetched; they have been reported to be carcinogenic, mutagenic and

teratogenic (Idowu et al. 2018). Sixteen of the PAHs have been listed as priority PAHs by the United States Environmental Protection Agency (US EPA). The sixteen priority PAHs include Naphthalene (Naph), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pry), Benz[a]anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (Bbf), Benzo[k]fluoranthene (Bkf), Benzo[a]pyrene (BaP), Indeno[1,2,3-cd]pyrene (Ip), Dibenzo[a,h]anthracene (DBahA), Benzo[ghi]perylene (B(g,h,i)P) (REFs). In addition, seven of the US EPA priority PAHs have been listed as carcinogens Abdel-Shaf and Mansour, 2015. The carcinogenic PAHs include benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1, 2, 3 cd]pyrene, and Dibenzo(a,h)anthracene.

In Nigeria, studies on environmental pollution by energy production systems abound. Elevated levels of criteria air pollutants were found to be associated with gas-fired thermal plants for electricity generation Sonibare, 2010. Emissions from heavy-duty trucks impacted the ambient air at a haulage vehicle park negatively (Lala et al., 2019). A review of anthropogenic air emissions in Nigeria showed that energy production systems stood out as significant drivers (Okedere et al. 2021). Also, some recent studies which focused on PAHs reported their elevated levels around industrial areas as well as regions of high vehicular activities (Fakinle et al., 2018b; Salaudeen et al., 2017). PAHs were also found to be associated with medical and municipal waste incinerators (Adesina et al., 2018 and Adesina et al., 2020). None of these studies examined the presence of PAHs in ambient air around a typical university powerhouse. Due to limited hours of electricity supply from the national grid, most universities in Nigeria run diesel-fueled back-up generators to meet the shortfall in their energy need. The present study investigated the ambient air

concentrations of PAHs near diesel-fueled generators run by Osun State University, Osogbo, Nigeria. This was intended to determine the presence and levels of carcinogenic PAHs around the power house. The study also determined the toxicity equivalent quotient for the individual PAHs and the entire priority PAHs.

METHODS AND MATERIALS

Description of the study area

The study area was near diesel - fueled electrical power generators owned by Osun State University, Osogbo, Nigeria. The university was established as the 30th State Government owned university and 80th

university in Nigeria in 2006, although actual operation began in 2007. Between 2007 and March 2021, the university operated diesel-fueled -electric power generators located on a plot of about 15.4 m x 30.8 m not too far from the administrative building. These generators were frequently deployed to meet the shortfall in the electric power supply to the university from the country’s national electricity grid. The generators were recently relocated to another piece of land (15.4 m x 53.9 m), about 400 m from the old site. Human activities are prevalent around the old and new sites of the generator whose geographic positions are described and summarized in Table 1.

Table 1: Geographic positions of sampling locations at the old and new generator sites

Sampling location	Item description	Geographic position
L1	Old generator site	07° 45’ 35.27”N and 04° 35’ 59.80 ”E
L2	Old generator site	07° 45’ 34.75”N and 04° 35’ 59.78 ”E
N1	New site for the generators close to mechanical engineering workshop	07° 45’ 34.78”N and 04° 36’ 0.77 ”E
N2	New site for the generators along walkway to Faculty of Environmental Sciences	07° 45’ 33.79”N and 04° 36’ 0.54 ”E
N3	Approach side to the new generator site	07° 45’ 33.98”N and 04° 36’ 0.19 ”E
N4	New site for the generators. Front of mechanical engineering workshop	07° 45’ 40.06”N and 04° 36’ 2.59 ”E
CP	Administrative block walkway	07° 46’ 22.10”N and 04° 35’ 45.56 ”E

Sampling and analytical procedures

Passive samplers made of polyurethane foams (PUF) were inserted between two stainless steel plates and mounted on an iron rod stand at about 1.5 meters above the ground. Three and five sampling locations were considered at the old and new sites, respectively and the PUF samplers were left at the sampling locations for 28 days. During this period, there were no activities at the old site, having relocated the generators earlier. The decision to place

samplers at the old site was to ascertain the impact of the previous activities at the site after. PUF passive samplers were also placed far away from the two sites as a form of control. The methods of extraction and the analytical procedures follow those reported in previous literatures.18-20 The PUF disks were withdrawn after 28 –days and subjected to 24- hour sohxlet extraction using dichloromethane to extract the PAHs from the PUFs. Cleaning up and elution were done with 5 g of silica gel column and

40 mL 1:1, DCM: Hexane followed by concentration in a rotary evaporator using a stream of nitrogen. Analysis of the samples was achieved with gas chromatography (Agilent 7890) with mass detector (Agilent 5975) that was done in selected ion monitoring mode and using electron impact ionization. The chromatographic column has a dimension of 30 m × 0.25 mm with internal diameter × 0.25 µm film thickness. The temperature program for the analytical procedure was set as: 90 °C (1.0 min), 30 °C/min, 250 °C, 4 °C/min, 330 °C (5 min). External standard method was used for the determination of PAHs in laboratory and field blanks. Pre-extraction procedure involved spiking of the samples with 25 mL of recovery standard (RS) which contained 20 ng of phenanthrene d₁₀ recovery ranged between 80 and 90%. Field blanks were lower than detection limit for all the investigated PAHs; no blank correction was carried out. The concentrations of PAHs in air were obtained by dividing the amount deposited on the PUF (µg) by air volume (m³). Global Atmospheric Passive Sampling (GAPS) network template was

used for the computation of the effective air volume. Duration of deployment, mean temperature, rate of sampling were the template input parameters (Adesinal et al., 2021; Pozo et al., 2013; Harner et al., 2013).

RESULTS AND DISCUSSION

Concentrations of priority paths

The concentrations of the sixteen priority PAHs in the ambient air of the old generator site are summarized in Table 2. All the sixteen priority PAHs were detected in the ambient air of the old generator site, an indication that the PAHs can exhibit a level of persistence in air. The predominant PAH around the old site of the generator was indeno [1,2,3-cd]pyrene with mean concentrations of 0.64 µg/m³ and 0.78 µg/m³ at sampling locations L1 and L2, respectively. The PAH with the least ambient air concentration around the old site was fluoranthene with an approximate mean value of 0.03 µg/m³. The PAHs levels were generally lower at the control site than at the two on-site locations.

Table 2: PAHs concentrations around the old site of the diesel-fueled power plant

PAH	Sampling Location (L1)			Sampling Location (L2)			Control
	S1	S2	Mean	S1	S2	Mean	Mean
Naph	0.0693	0.0729	0.0711	0.0724	0.0666	0.0695	0.0115
Ace	0.0564	0.0484	0.0524	0.0473	0.0559	0.0516	0.0168
Acy	0.0958	0.1062	0.101	0.0925	0.0895	0.091	0.0321
Flu	0.0504	0.0464	0.0484	0.0444	0.0508	0.476	0.024
Phe	0.0415	0.0389	0.0402	0.0411	0.0425	0.0418	0.012
Ant	0.4136	0.5228	0.4682	0.6017	0.5819	0.5918	0.206
Flt	0.0327	0.0291	0.0309	0.0345	0.0289	0.0317	0.0143
Pyr	0.0473	0.0525	0.0499	0.0398	0.0404	0.0401	0.021
BaA	0.4892	0.5468	0.518	0.5819	0.6221	0.602	0.24
Chr	0.264	0.2344	0.2492	0.2503	0.2913	0.2708	0.102
Bbf	0.4385	0.3587	0.3986	0.3973	0.4455	0.4214	0.23
Bkf	0.4385	0.3773	0.4079	0.4314	0.3968	0.4141	0.21
BaP	0.4274	0.4814	0.4544	0.5467	0.5845	0.5656	0.37
Ip	0.6194	0.6622	0.6408	0.8028	0.7636	0.7832	0.453
DB(ah)A	0.2914	0.3482	0.3198	0.3388	0.3816	0.3602	0.1901
B(g,h,i)P	0.6174	0.5312	0.5743	0.6605	0.5909	0.6257	0.1502
Total			4.4251			5.4365	

The total priority PAHs concentrations at the sampling locations L1 and L2 on the old site were 4.43 $\mu\text{g}/\text{m}^3$ and 5.44 $\mu\text{g}/\text{m}^3$, respectively. The obtained Σ priority PAHs at the old site of the diesel-fueled generators exceeded the regulatory limit of 10 $\mu\text{g}/\text{m}^3$ set for ambient air total PAHs in China and European countries (Pozo et al., 2013; Harner et al., 2013).. It is customary to classify PAHs as low molecular weight (LMW) and high molecular weight (HMW) depending on the number of benzene rings present. Usually, PAHs having 2 to 3 and 4-6 benzene rings are labeled LMW and HMW, respectively. LMW and HMW PAHs constituted 17.7% and 82.3% of the PAHs detected at L1 while at L2, the percentage detections were LMW (24.3%) and HMW (75.7%), respectively. Hence, the measured Σ priority PAHs were dominated by HMW PAHs, which agrees with previous studies which indicated that predominance of HMW over LMW PAHs was a pointer to combustion sources (Wang, 2018; Onyema et al., 2012; Oyewo, et al., 2022).

Seven of the priority PAHs have been reported to be carcinogenic and these include benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzofluoranthene, benzo[k] fluoranthene, benzo[a] pyrene, indeno[1, 2, 3-cd]pyrene, and Dibenzo(a,h)anthracene. The total concentrations of the carcinogenic PAHs (Σ carcinogenic PAHs) were approximately 2.98 $\mu\text{g}/\text{m}^3$ and 3.42 $\mu\text{g}/\text{m}^3$ at L1 and L2 respectively. The Σ carcinogenic PAHs obtained in this study are well above the thresh-hold limit set for ambient air by the US EPA.

The obtained concentrations of PAHs at the present location of the generators are summarized in Table 3. Similar to what was observed at the old site, all sixteen priority PAHs were detected at the current location of the power plant. However, based on the obtained mean concentrations, the predominant and the least PAHs around the present power plant site were

benzo[a]anthracene and acenaphthene, with mean concentrations of 2.83 $\mu\text{g}/\text{m}^3$ and 0.12 $\mu\text{g}/\text{m}^3$, respectively. The Σ priority PAHs ($\mu\text{g}/\text{m}^3$) at the different sampling locations in the vicinity of the new site of the power plant were N2 (5.35) > N4 (4.83) > N3 (4.60) > N (4.47). These results compare favourably with the ambient concentrations of PAHs around the previous location of the power plant, which ranged between 4.43 $\mu\text{g}/\text{m}^3$ and 5.44 $\mu\text{g}/\text{m}^3$. Similarly to what was observed for the old site of generators, the HMW PAHs dominated the measured PAHs. The percent HMW PAHs at N1 was 88.7%, while at N2, N3, and N4, it was 89%. The Σ carcinogenic PAHs ($\mu\text{g}/\text{m}^3$) at the present location of the generators were in the order N2 (4.17) > N4 (3.76) > N3 (3.58) > N (3.35). The Σ carcinogenic PAHs ($\mu\text{g}/\text{m}^3$) were thus greater at the present location of the generators than at the former site.

Table 3: PAH levels at the new locations of the generators

PAHS	N1	N2	N3	N4
Naph	0.0689	0.0537	0.06	0.0474
Ace	0.0326	0.0242	0.0266	0.0316
Acy	0.0778	0.0994	0.0616	0.0745
Flu	0.0269	0.0358	0.031	0.0298
Phe	0.0709	0.0676	0.0866	0.0731
Ant	0.2297	0.3346	0.2423	0.2771
Flt	0.093	0.0724	0.0621	0.0742
Pyr	0.0654	0.1121	0.0684	0.0801
BaA	0.7032	0.8474	0.5853	0.692
Chr	0.7385	0.6035	0.6822	0.6303
Bbf	0.3513	0.4901	0.4159	0.4651
Bkf	0.294	0.4356	0.3223	0.3356
BaP	0.4684	0.8304	0.5201	0.621
Ip	0.4148	0.4398	0.7148	0.5502
DB(ah)A	0.3775	0.5275	0.34	0.4511
B(g,h,i)P	0.4555	0.3736	0.3803	0.4003

The levels of PAHs obtained in this study were compared with their ambient air levels reported for other combustion sources. The levels of PAHs (Σ priority PAHs) obtained in this study are more than those reported

for ambient air near the medical wastes' incinerator operated by a teaching hospital in Nigeria (Adesina et al., 2018). In another study, it was reported that the Σ priority PAHs for places around a municipal waste dumpsite that is frequently subjected to open burning ranged between $3.29 \mu\text{g}/\text{m}^3$ and $4.02 \mu\text{g}/\text{m}^3$ (Adesina et al., 2020). This is lower than the range of $4.47 - 5.35 \mu\text{g}/\text{m}^3$ obtained in this study for ambient air around the diesel-fueled power plant being operated by Osun State University.

Toxicity equivalence (TEQ)

The extent of toxicity of the individual PAH is commonly expressed in terms of toxicity equivalence (TEQ). This is usually expressed as a product of the toxicity equivalent factor (TEF) and the measured

concentration. In discussing the toxicity of PAHs, Benzo[a]Pyrene are considered the reference PAH and is assigned a TEF of 1.00. The TEQs of the sixteen priority PAHs around the old site of the power plant are depicted in Table 3. Benzo[a]Pyrene had the highest value for TEQ and is the most significant contributor to the total toxicity equivalence (TTEQ) with 65.3% and 67.4% share at L1 and L2, respectively. Apart from Benzo[a]pyrene which had the highest TEQ, indeno[1,2,3-cd]pyrene, benzo[a]anthracene, benzo[k]fluoranthene, and benzo[b]fluoranthene were the other PAHs with a fair share of the TTEQ. The TTEQ at the two sampling locations chosen for investigation around the old site of the power house were of the trend L2 (0.839) > L1 (0.696). μ

Table 3: TEQs and TTEQs of priority PAHs around the old and new sites (g/m^3)

PAHs	TEF	TEQ (L1)	TEQ (L2)	TEQ (N1)	TEQ (N2)	TEQ (N3)	TEQ (N4)
Naph	0.01 ^b	0.0711 ^a	0.0695 ^a	0.0689 ^a	0.0537 ^a	0.0600 ^a	0.0474 ^a
Ace	0.001 ^b	0.0524 ^a	0.0516 ^a	0.0326 ^a	0.0242 ^a	0.0266 ^a	0.0316 ^a
Acy	0.001 ^b	0.1010 ^a	0.0910 ^a	0.0778 ^a	0.0994 ^a	0.0616 ^a	0.0745 ^a
Flu	0.001 ^b	0.0484 ^a	0.4760 ^a	0.0269 ^a	0.0358 ^a	0.0310 ^a	0.0298 ^a
Phe	0.001 ^b	0.0402 ^a	0.0418 ^a	0.0709 ^a	0.0676 ^a	0.0866 ^a	0.0731 ^a
Ant	0.01 ^b	0.0047 ^a	0.0059 ^a	0.0023 ^a	0.0033 ^a	0.0024 ^a	0.0028 ^a
Flt	0.001 ^b	0.0309 ^a	0.0317 ^a	0.0930 ^a	0.0724 ^a	0.0621 ^a	0.0742 ^a
Pyr	0.001 ^b	0.0499 ^a	0.0401 ^a	0.0654 ^a	0.1121 ^a	0.0684 ^a	0.0801 ^a
BaA	0.1 ^b	0.0518 ^b	0.0602 ^b	0.0703 ^b	0.0847 ^b	0.0585 ^b	0.0692 ^b
Chr	0.01 ^b	0.0025 ^b	0.0027 ^b	0.0074 ^b	0.0060 ^b	0.0068 ^b	0.0063 ^b
Bbf	0.1 ^b	0.0399 ^b	0.0421 ^b	0.0351 ^b	0.0490 ^b	0.0416 ^b	0.0465 ^b
Bkf	0.1 ^b	0.0408 ^b	0.0414 ^b	0.0294 ^b	0.0436 ^b	0.0322 ^b	0.0336 ^b
BaP	1 ^b	0.4544 ^b	0.5656 ^b	0.4684 ^b	0.8304	0.5201 ^b	0.6210 ^b
Ip	0.1 ^b	0.0641 ^b	0.0783 ^b	0.0415 ^b	0.0440	0.0715 ^b	0.0550 ^b
DB(ah)A	0.1 ^b	0.0320 ^b	0.0360 ^b	0.0378 ^b	0.0528	0.0340 ^b	0.0451 ^b
B(ghi)P	0.01 ^b	0.0057 ^b	0.0063 ^b	0.0046 ^b	0.0037	0.0038 ^b	0.0040 ^b
TTEQ		0.6962 ^b	0.8394 ^b	0.6972 ^b	1.1180	0.7714 ^b	0.8839 ^b

Where ^a and ^b are units in micrometer ($\mu\text{g}/\text{m}^3$) and millimeter (mg/m^3), respectively.

The TTEQs of PAHs in ambient air around the new location of the power plant were of the trend N2 (1.118) > N4 (0.8839) > N3 (0.7714) > N1 (0.6972) as shown in Table 3. Similar to the TEQs obtained for the individual PAH around the old site, BaP

was also observed to be the most significant contributor to TTEQs around the new site, accounting for 67.2%, 74.3%, 67.4% and, 70.3% of the obtained TTEQ at N1, N2, N3, and N4 respectively.

Generally, the toxicity of PAHs is usually expressed in terms of the concentrations of BaP. The permissible limit set for ambient air BaP by the European Union (EU) and World Health Organization (WHO) is $1 \mu\text{g}/\text{m}^3$, while the United Kingdom has a threshold limit of $0.25 \mu\text{g}/\text{m}^3$ (Ravindra et al., 2008; Vardoulakis et al., 2008; Ana et al., 2012). The ambient air BaPs at both sites (old and new) of the power plant (this study) are higher than the recommended limits, and the atmosphere is therefore considered unsafe due to the higher levels of carcinogenic PAHs above the permissible limits.

Diagnostic ratios

PAHs diagnostic ratios are binary ratios commonly used to infer the likely nature of a source of PAHs (Ravindra et al., 2009; Okedere et al., 2022). They are established ratios that help identify the sources of PAHs measured at a given receptor

location. Some reported PAHs diagnostic ratios, indicators of diesel-related sources and the obtained ratios in this study are presented in Table 4. When the diagnostic ratio $[\text{Ind}/(\text{Ind}+\text{B}(\text{ghi})\text{P})]$ was applied to the measured PAHs, the ratios (R) were in the range $0.35 \leq R \leq 0.7$ at L1 (0.5274), L2 (0.5559), N1 (0.4766), N2 (0.5407), N3(0.6527) and N4 (0.5789). The obtained results agree with the diagnostic ratio $\text{BbF}/\text{BkF} > 0.5$ at all the locations, which indicated that the measured PAHs are traceable to diesel combustion. The last diagnostic ratio applied was $\text{CPAHs}/\Sigma\text{PAHs} \approx 1.0$. The CPAHs is the sum of major non-alkylated PAHs while ΣPAHs is the total concentration of the priority PAHs. The obtained ratios for CPAHs/ ΣPAHs in this study ranged from 0.76 to 0.81. They are large enough and rounding them off to the nearest whole number will approximate 1.0. The PAHs diagnostic ratios, therefore, affirmed that the measured PAHs were from diesel-related sources.

Table 4: PAHs diagnostic ratios

Diagnostic ratios for diesel related sources	Diagnostic ratios obtained in this Study					
	L1	L2	N1	N2	N3	N4
$R = \text{Ind}/[\text{Ind} + \text{B}(\text{ghi})\text{P}] ; 0.35 \leq R \leq 0.7$	0.5274	0.5559	0.4766	0.5407	0.6527	0.5789
$R = [\text{BbF}/\text{BkF}] > 0.5$	0.9772	1.0176	1.1949	1.1251	1.2904	1.3859
$R = [\text{CPAHs}/\text{PAHs}] \approx 1.0$	0.7551	0.7724	0.7873	0.7795	0.8088	0.7871

CONCLUSION

The levels of priority PAHs in the vicinity of diesel – fueled electric power generators operated by Osun State University, Osogbo, Nigeria have been investigated in this study with a view to ascertain the quality of ambient air around the power plant. All sixteen United State Environmental Protection (US EPA) priority polycyclic aromatic hydrocarbons (PAHs) were detected in ambient air around the generators. The higher molecular weight PAHs were dominant in the analyzed samples. The PAHs levels obtained in this study were above the permissible limits set by

the US EPA and European Union for total ambient air PAHs. The presence of carcinogenic PAHs at levels above the permissible limit calls for concern. Being a university campus with human activities around the locations of the generators, the study concluded that both the old and new locations of the generators are not appropriate. A much more remote site could be considered for the generators to minimize the chance of human exposure. Further studies should be conducted to ascertain the PAH levels in the same area since the soil is always considered the final sink for most pollutants.

Disclosure Statement

The authors declare no conflict of interest.

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