

## Investigations into the transition to sustainable alternative fuels in a South African underground platinum mine

G. Geldenhuys<sup>a,b</sup>, M. Wattrus<sup>c</sup>, M. Fox<sup>b</sup>, P.B.C. Forbes<sup>a,\*</sup>

<sup>a</sup> Department of Chemistry, Faculty of Natural and Agricultural Sciences, University of Pretoria, Pretoria 0002, South Africa

<sup>b</sup> Impala Platinum Ltd, 2 Fricker Road, Illovo 2196, South Africa

<sup>c</sup> Sasol, 50 Katherine Street, Sandton, 2196, South Africa

### ARTICLE INFO

#### Keywords:

Polycyclic aromatic hydrocarbons  
Diesel exhaust emissions  
Biofuel  
Sustainability

### ABSTRACT

Adverse environmental impacts associated with the use of fossil fuels and the over-dependence thereon has made energy security and sustainability a critical issue worldwide particularly for key energy intensive economic sectors which are heavily dependent on diesel. We thus investigated the feasibility of a transition to two different alternative fuels namely, rapeseed methyl ester (RME) biodiesel and gas-to-liquid fuel (GTL), in the platinum mining industry in South Africa. Load haul dump vehicles are the most abundant workhorses underground and were the selected vehicles to test alternative fuels at 100% without any engine modification. Potential reduction of harmful unregulated polycyclic aromatic hydrocarbon (PAH) emissions was the focus of the research due to their adverse impacts on the environment, human health and engine operations. Quantitative collection of gas and particle phase PAHs was made possible using portable denuder devices followed by analysis by two-dimensional gas chromatography coupled to time-of-flight mass spectrometry. Results showed that total PAH emissions from a high idling vehicle decreased dramatically when diesel was substituted with both biofuels (total gas phase PAH concentrations of 34; 14 and 9  $\mu\text{g m}^{-3}$  for diesel, GTL and RME, respectively) and no substantial hindrance on engine performance was reported. This novel sector specific study on unmodified heavy duty working vehicles can potentially translate into a real-world, immediate solution, as not only would the selected biofuels be able to directly replace diesel, but both have high potential of being locally produced in South Africa and assist in the promotion of a circular economy.

### Introduction

Environmental protection and the search for cleaner, more sustainable fuels are two of the most important concerns modern society is facing today. The over-dependence on oil and natural gas, the intensifying demand for energy, as well as fossil fuel shortages have made energy security a critical issue worldwide [1]. Although the economic impact may be negative, the adverse environmental impacts associated with the use of fossil fuels are far more extensive and will influence the planet and all future generations to come.

The projected negative impacts of climate change as well as the volatility of oil supply are forcing governments to search for alternative options. Several countries have already developed regulatory frameworks to allow the approval of biofuels and biofuel blends to meet transportation requirements and thereby reduce the reliance on conventional petroleum imports to help mitigate the impacts of volatile oil

prices and reduce carbon emissions. Ebadian et al. for example, summarised biofuel mandates and their effectiveness on biofuel markets in numerous countries [2].

In addition to the extensive list of advantages listed in literature, biofuel generation and integration will encourage new entrepreneurs and boost the South African (SA) economy, whilst simultaneously increasing economic activity internationally [3]. The use of biofuels will not only reduce SA's energy dependence on imported fuels but it can lead to substantial reductions in air emissions of potentially harmful substances and greenhouse gases (GHGs), and thus to healthier, sustainable development.

This potential has been acknowledged by government and in 2005, the then Department of Minerals and Energy (DME) (now the Department of Minerals, Resources and Energy (DRME)) proposed the development of a biofuels industry to Cabinet who went on to appoint and approve an inter-departmental Biofuels Task Team (BTT) [4]. The BTT

\* Corresponding author.

E-mail address: [patricia.forbes@up.ac.za](mailto:patricia.forbes@up.ac.za) (P.B.C. Forbes).

<https://doi.org/10.1016/j.ref.2023.100500>

Received 21 July 2023; Received in revised form 7 October 2023; Accepted 11 October 2023

Available online 12 October 2023

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drafted the Biofuels Industrial Strategy (BIS) which provided a five-year pilot phase from 2008 to 2013 to initially introduce a 2 % penetration level of biofuels in the national transport fuels pool (petrol and diesel), whilst monitoring the resultant socio-economic benefits and adverse consequences. The Green Transport Strategy of South Africa [5] identifies biofuels as one of the clean transport fuels for the transition towards a lower carbon transport future of the country.

One of the biggest industries contributing to the South African economy, contributing 7.53 % to the gross domestic product (GDP) [6], is the platinum mining industry, which is currently heavily dependent on fossil fuels. Critical mining operations include the extensive use of trackless mobile diesel machinery (TMM) including: load haul dump vehicles (LHDs), drill rigs, dump trucks, utility vehicles (UVs) and cherry pickers. Combustion emissions from these sources contribute to gas and particulate air pollutant levels, including those of polycyclic aromatic hydrocarbons (PAHs) which are semi-volatile organic compounds (SVOCs) that exist in both gas and particulate phases. In literature, reductions in regulated pollutant emissions when using biofuels or blends thereof have been well studied, whereas unregulated emissions such as PAHs lack extensive research. These compounds are not only a cause for concern from an environmental standpoint, as they are ubiquitous environmental contaminants, but also from a human health perspective, especially due to the confined working environments underground which results in an increased risk of occupational exposure to harmful and carcinogenic pollutants [7]. PAHs can also be toxic and hazardous to engine components as the condensation and accumulation of PAHs on the exhaust valve stem causes wet stacking or engine failure [8]. In the quest for greener fuels, it is crucial to examine levels of unregulated emissions such as PAHs which are currently not controlled by the engine manufacturers nor regulated by the government agencies but play a crucial role in determining fuel use feasibility.

A comparative assessment of gas and particle PAH emissions from two diverse diesel generator engines using 100% biodiesel and diesel fuel was conducted by Yilmaz & Donaldson. Their results indicated that the use of biodiesel in both engines reduced total PAH emissions by 48.02% – 49.36% and PAH toxicity by 83.49% – 84.8% as compared to diesel fuel, which was attributed to the aromatic content of the base fuel [9]. In another study where the fuel effects on PAH formation, toxicity and regulated emissions were investigated from an indirect-injected, four-cylinder diesel generator engine, the authors reported that biodiesel and biodiesel-alcohol blends (propanol, n-butanol, and 1-pentanol at 5 %, 20 % and 35 %) significantly decreased formation of PAHs and associated toxicity with the addition of 5 % alcohol to biodiesel further decreasing total PAH emissions when compared to pure diesel [8].

A transition to alternative fuels, in these critical mining operations, can lead to a reduction in emissions of harmful pollutants, including PAHs, and greenhouse gases, but the transition must be accompanied by a comprehensive risk management strategy, as there are many other aspects that need to be considered on a national level such as the type, source and availability of an alternative fuel, as well as the cost of implementation. Other clean energy solutions such as hydro, solar and nuclear energy are limited, as they do not produce liquid fuel that is needed in transportation [10]. Fuel cell technology offers many advantages that can assist in solving safety, health, operational efficiency and productivity in the mining industry, as well as zero environmental emissions. However, currently the growth of hydrogen use is limited by the lack of hydrogen infrastructure, perceptions around hydrogen safety and sustainable development of a hydrogen economy [11]. One of the largest contributing factors is also the initial capital expenditure required, as diesel engines would have to be entirely replaced which may provide a long-term solution but cannot be implemented immediately.

The overarching goal of this research was to determine the broad feasibility of a transition from fossil fuels to the use of biofuels in the South African underground platinum mining industry and to assess

whether such a transition could potentially serve as a sector-based solution to the energy crisis in developing countries. The experimental portion of this study tested the use of biofuels, using 100% unblended biofuel as a direct replacement for diesel in engines used underground, and thus provides new data to fill in the gaps on this subject in the literature. To achieve this, we investigated an LHD engine performance and emissions when tested with 100% rapeseed methyl ester biodiesel (RME) and 100% synthetic gas-to-liquid (GTL) respectively. The effect of these greener fuels on the emission of unregulated polycyclic aromatic hydrocarbon compounds produced by the diesel engine was investigated due to their adverse effects on the environment, human health and engine components. The investigation encompassed two very different biofuels which were used as a direct diesel replacement in real-world operating diesel engines in an underground mining environment for the first time. Further, unregulated emissions were monitored through the determination of the concentrations of 15 priority PAHs.

These selected test fuels have the potential of being generated locally with sustainable feedstocks for first or second generation biofuel such as from sugar cane, sunflower oil ester or waste biomass. The study was thus conducted with consideration of both the present and the prospective energy sector in South Africa and the growing demand for healthier, more sustainable development. The findings of this study may thus aid in the determination of a strategy that could be implemented relatively quickly and cost efficiently in a manner that paves the way to biofuel industrialisation in South Africa.

## Materials and methods

To determine the feasibility of a transition from fossil fuel to the use of greener fuel in the platinum mining industry, three indicators were assessed namely:

- Reduction in air pollutant emissions as assessed by an underground sampling campaign with focus on unregulated PAHs
- Functional performance of the trackless mobile machinery
- Environmental and socio-economic factors

The methods adopted to evaluate each indicator are described in this section.

### *Reduction in air pollutant emissions as assessed by a sampling campaign*

#### *Sampling Site*

The sampling campaign took place in the underground workings of a platinum mine in the North West Province of SA. The test LHD vehicle was positioned upwind from any mining activity to prevent other sources of emissions affecting the measurements and a more detailed description of the sampling site is given in Section S1 of the SI, whilst details of the samples taken during the sampling campaign are presented in Table S1.

#### *Test fuels*

Three test fuels were evaluated:

- (1) Petroleum-based diesel containing 50 ppm sulfur (pure diesel (PD)), representing the reference diesel used in the workplace (status quo),
- (2) Paraffinic fuel which was EN15940 compliant (GTL, ORYX, Qatar),
- (3) Biofuel which was compliant to EN14214, stabilized with oxidation stabilizer (RME, SBE BioEnergie, Germany).

Properties of the diesel, RME and GTL fuel used in the underground sampling campaign are provided in Table S2.

#### *Test engine specifications and operating procedure*

One designated LHD (see Table S3 for engine specifications and Figure S1 for a photograph of a LHD) was used throughout the sampling

campaign. The LHD was mid-service interval and was a good representative of the average fleet in use in South African underground mines. A more detailed account of the LHD operating procedure can be found under Section S2 of the SI.

#### PAH sampling methodology

Air samples were collected underground with personal sampling pumps attached to portable denuder devices (Fig. 1) that were comprised of two multi-channel silicone rubber traps that were separated by a quartz fibre filter (QFF). These devices allowed for simultaneous gas and particulate PAH sampling and have been validated in numerous studies [12–17]. The denuder sampler methodology is described in detail in Section S3 of the SI.

#### Test procedure

The LHD was operated in two modes namely high idle mode (HI) and a test cycle mode (TC). HI mode was characterised by full acceleration with no throttle and TC was a load test whereby the LHD bucket lifted a fixed mass (one LHD tyre) to mimic the LHD in operation. More information on underground LHD operation can be found in Wattrus *et al.* [18] and under Section S4 in the SI.

#### Analytical method

Offline analysis of individual denuder components was performed using thermal desorption coupled to two-dimensional gas chromatography with time-of-flight mass spectrometric detection (TD-GCxGC-ToFMS). Calibration was performed using a certified standard PAH mix solution (Supelco, St Louis, MO), that contained 15 priority PAHs as listed in Table S4. Additional details on the instrumental and statistical analysis, calibration procedure and quality assurance are provided in the SI under Sections S5–S8.

#### Functional performance of the trackless mobile machinery

Evaluation of this indicator was performed by conducting a literature review pertaining to the effects that biofuels can have on engine performance which could not be experimentally measured due to lack of resources. Physical observations were, however, made during the sampling campaign by the LHD operator, researchers and selected mine staff regarding engine performance, smoke opacity and volume, and visible

particulate matter (soot) when the LHD was operated on pure diesel, RME and GTL, respectively. The literature findings as well as feedback obtained from experimental observations were evaluated and used to draw conclusions on the engine functional performance indicator.

#### Environmental and socio-economic factors

Environmental, economic, and social impacts pertaining to the use of diesel, biodiesel and GTL/ biomass-to-liquid (BTL), respectively were reviewed and defined followed by the evaluation of this indicator by comparing and tabulating these factors for three scenarios namely, the current use of diesel, prospective use of greener alternative BTL and prospective use of greener alternative biodiesel.

## Results and discussion

#### Reduction in air emissions as assessed by a sampling campaign

A recent comprehensive review on the effects of biofuel on engine performance and emissions verifies that all biodiesels and their blends have demonstrated the ability to improve engine performance, as well as the ability to reduce pollutants such carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), hydrocarbons (HCs), particulate matter (PM) and under various operating conditions even nitrogen oxide (NO<sub>x</sub>), but the vital importance of understanding the combustion properties of fuels is emphasised [19]. The use of biodiesel underground can thus provide numerous potential benefits to the mining industry pertaining to sustainability and improvements to human and environmental health. Additionally, biofuels are non-toxic, biodegradable and sulfur free and as a result, no sulphates are formed at high gas temperatures and heavy loads as is the case with heavy duty fleets [20] such as those used in underground mines.

Moon *et al.* characterised emissions from diesel, GTL, and biodiesel-blended fuels that were used in a diesel engine, and found noticeable decreases in CO (16–52 %) and total hydrocarbon (THC) (22–56 %) emissions for biodiesel-GTL blends whereas NO<sub>x</sub> emissions increased by up to 12 % when compared to diesel due to increased oxygen content and higher temperatures in the combustion chamber [21]. The higher oxygen content for biofuel improves combustion quality but the increased temperatures favour the formation of NO<sub>x</sub> through the

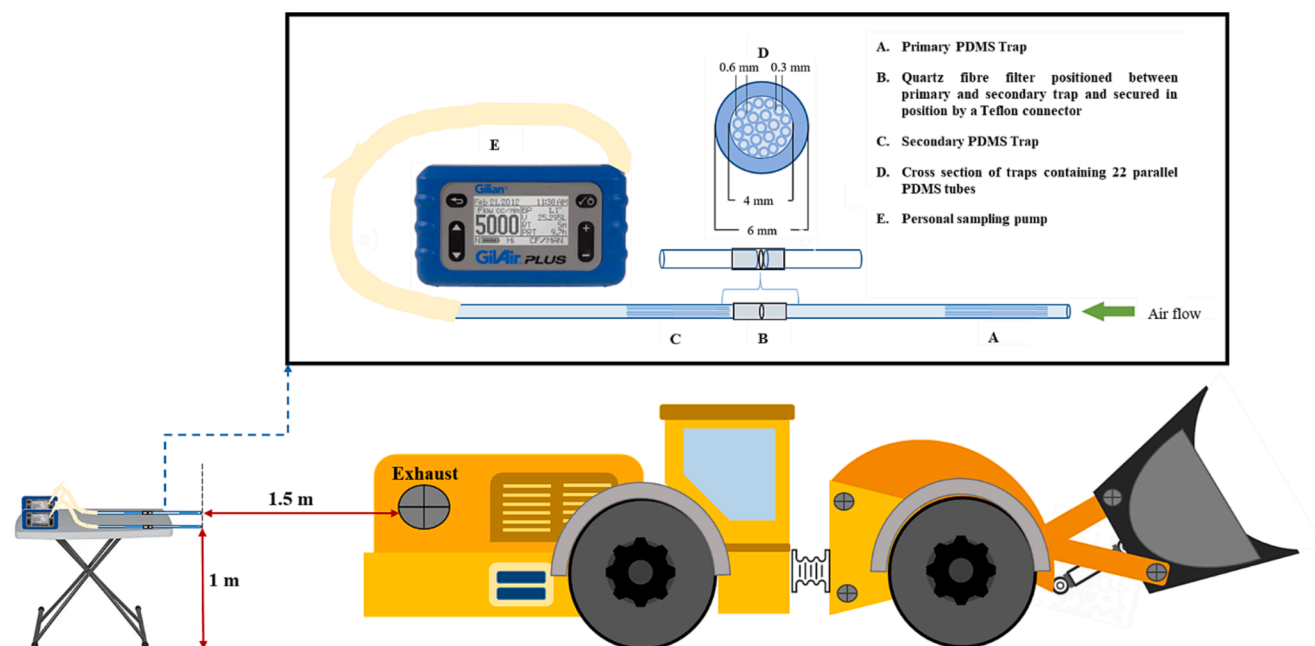


Fig. 1. Schematic representation of the PDMS denuder device used for PAH sampling and the position relative the LHD tailpipe.

Zel'dovich mechanism [22]. The emissions of  $\text{NO}_x$  from combustion can be mitigated by internal measures, such as exhaust gas recirculation (EGR) whereby emissions are controlled by reducing the oxygen concentration inside the cylinder and thereby reducing the flame temperature of the charge mixture inside the combustion chamber [23]. EGR with fuel lean conditions and water injection, combined with exhaust after-treatment employing techniques such as lean  $\text{NO}_x$  traps (LNT), diesel particulate filter (DPF) and selective catalytic reduction (SCR), have shown to be efficient in dual-fuel applications [22,24].

This study was carried out with the aim of assessing the impacted environment underground due to combustion emissions from LHD vehicles operating on various fuels. PAHs were the target analytes due to their human and environmental toxicity as well as the paucity of information on gas and particle phase characterisation of these compounds in the underground environment [15]. PAHs can generally originate from three different sources including pyrosynthesis during combustion, from the original unburned fuel or the lubrication oil respectively, and lastly formation by the hydrogen abstraction- $\text{C}_2\text{H}_2$  addition (HACA) mechanism [25,26]. Unsubstituted PAHs are prevalently formed by the HACA mechanism in combustion engines and the number of rings formed during this process is dependent on the combustion temperature and conditions [27].

The sampling campaign results presented in Fig. 2 revealed a clear difference in total PAH concentrations arising from exhaust emissions between the different fuels in both high idle and test cycle mode. It is evident that the PAH concentrations from the combusted diesel fuel are substantially higher than the emissions from the other fuels. Combustion of the diesel fuel produced the highest concentration of PAHs on the primary trap with a value of  $19 \mu\text{g m}^{-3}$  followed by the GTL fuel at  $6 \mu\text{g m}^{-3}$  and then the RME, which showed the lowest PAH concentrations which were almost 10-fold lower than that of the diesel fuel. These values showed the same trend but were higher than the PAH concentrations found in the study by Yilmaz et al. who reported total PAH concentrations of  $4.73$  and  $2.47 \mu\text{g m}^{-3}$  for diesel and unsaturated fatty acid methyl ester biodiesel emissions, respectively, which indicated a 48 % reduction [8]. The test engine age, size and condition would have substantially contributed to the lower emissions. This highlights the

need for the current study which provides emission data for heavy duty diesel engines under real-world usage scenarios.

The lower PAH concentrations from the use of GTL and RME fuels were expected as the biofuel does not contain aromatic hydrocarbons and the GTL aromatic content is greatly reduced when compared to crude derived diesel. RME is generated by transesterification of rapeseed oil and is primarily composed of fatty acid methyl esters (FAMES) with different length alkyl chains. This type of fuel does not contain aromatic compounds therefore no PAHs will result from unburned fuel, thus lower emissions of PAHs compared to diesel is expected and this was what was reported in numerous other studies [8,9,28–31].

These findings are in agreement with Yilmaz and Donaldson who concluded that PAH emissions and toxicity strongly depend on both the aromatic content of the fuel and the combustion processes and found that aromatic-free biodiesel significantly decreased PAH emissions from two different engines by 48%–49% as compared to diesel fuel. They also reported that benzo[a]pyrene, having the greatest carcinogenic threat, was eliminated in both engines as a result of using 100% biodiesel [9]. Similarly, the effects of biodiesel and diesel mixtures containing various blends n-propanol, n-butanol, and n-pentanol on PAH emissions were compared and the results showed that there was a significant reduction in PAH emissions by using alcohol at a high mixing ratio of 35% which also had positive effects on the engine's operating performance and the environment [8].

In a study whereby only PM was considered, a total of 17 PAHs were identified and quantified from the PM gathered from a Euro 4 compliant engine exhaust, equipped with an oxidation catalyst (DOC) and with a cooled EGR system, as the palm oil biodiesel concentration was increased. The 20% blend exhibited a reduction of around 50% in PAHs in comparison with the 10% blend, while the 100% blend reduced PAH emissions by 90%. It was noted that PAH reductions were more pronounced for heavier 5-6 ring PAHs [32]. In a recent study, 8 PAHs, 27 nitro-PAHs, and 6 quinones (oxy-PAHs) bound to combustion particles were determined by GC-MS in samples obtained from a 4-stroke, direct injection, turbocharged engine in a dynamometer study. Low molecular weight PAHs were mainly bound to PM particles emitted by a ternary blend of 90% v/v diesel, 7% v/v biodiesel and 3% v/v ethanol [33].

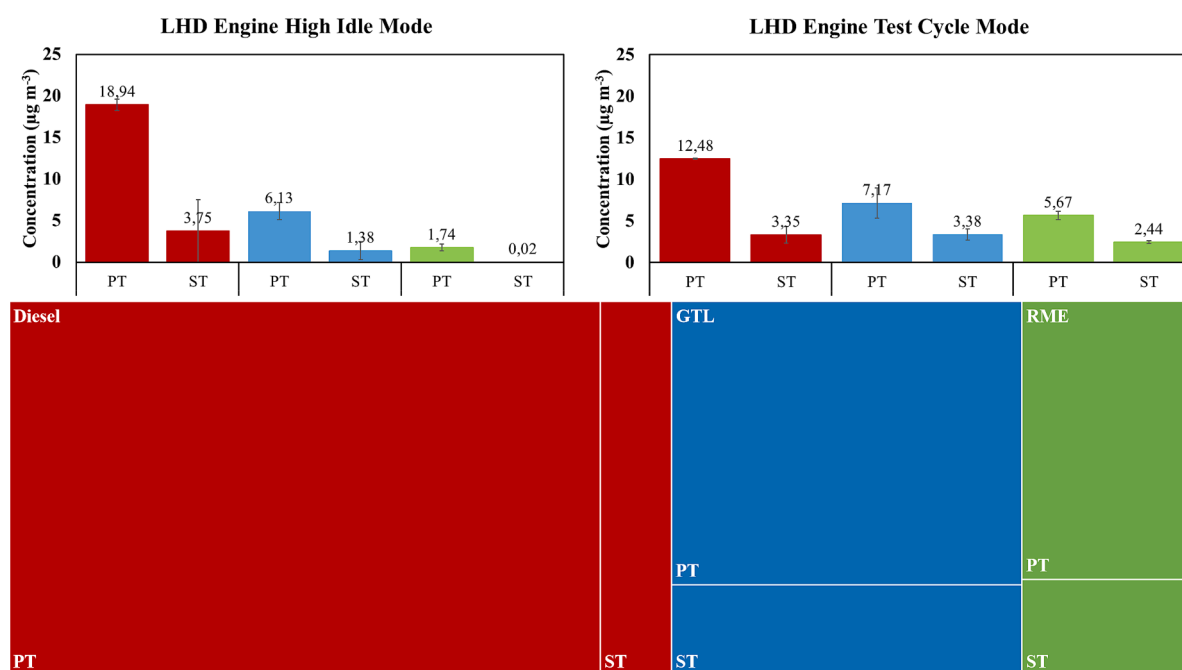


Fig. 2. (Top left) Total PAH concentrations on the primary and secondary trap for the LHD engine exhaust samples during high idle mode using diesel, RME and GTL fuels. (Top right) Total PAH concentrations on the primary and secondary trap for the LHD engine exhaust samples during test cycle mode using diesel, RME and GTL fuels and (bottom) hierarchical tree map chart of total PAH concentrations for different tested fuels during both cycles.

Only particulate sampling was done in these studies and it must be noted that the lack of gas phase sampling would result in an underestimation of total emission of PAHs.

Although not fuel derived, PAH emissions cannot be completely abated as they are still generated during combustion or through pyro-synthesis irrespective of PAH content in the fuel [26]. GTL is primarily composed of long-chain alkanes and is expected to emit lower amounts of particulate matter (PM) and lower amounts of PAHs compared to fossil fuel combustion. Although GTL uses natural gas as a feedstock, an alternative to the synthetic GTL offers a more carbon neutral pathway namely BTL fuel, which is produced in the same way as the GTL but from organic waste sources. The Fischer-Tropsch process is used to produce GTL from natural gas that results in a purely paraffinic liquid fuel that contains no aromatic compounds and can be used in conventional diesel engines without any modifications to the hardware [21]. Another important advantage of using biofuels, specifically biodiesel, in conventional diesel engines is the higher oxygen content due to the fatty acids that result in more complete combustion and thus lower emissions of harmful compounds, such PAHs [20,29–31].

The lower PM concentrations during combustion of biofuels would also result in lower PAH concentrations as these parameters have been positively correlated in other studies [33–35]. The PAHs that were detected upon biofuel combustion in this study can therefore be attributed to the pyrolysis products formed during combustion. The test cycle PAH concentrations followed the same trend, however the most noticeable difference was the higher values for the RME samples when the engine was under load, resulting in a lower ratio between the diesel and RME PAH concentrations.

The downwind samples reveal that the total PAH concentrations (sum of PT and ST) for the diesel, GTL and RME fuels during high idle mode were 23, 8 and  $2 \mu\text{g m}^{-3}$  respectively. The downwind samples are representative of the fresh exhaust emissions implying that the PAHs remained in the gas phase and no particle association occurred during the sampling period. This was consistent with other studies which found that the aromatic ring distribution of 100% biofuel exhaust consisted of 50–53% two, 32–34% three and 15–16% four aromatic rings which are predominantly associated with the gas phase and no higher-ring PAHs were found [9]. Tsai et al. also confirmed that low molecular weight PAHs were the most abundant PAHs that were emitted by diesel engines operated with different biodiesel blends [36].

The tree map in Fig. 2 displays the PAH concentration data for the different fuels in a hierarchical structure and it is evident that the PAH concentrations from the combusted diesel fuel are substantially higher than the PAH concentrations from the other fuels with the green section representing the RME biodiesel.

Based on these results it can be concluded that in addition to the reduction in known and regulated pollutants, such as PM, HCs, CO, and  $\text{NO}_x$  [37], the substitution to biofuels will also lead to a substantial reduction in toxic PAH emissions. A discussion of other chemical features contributing to the variance between PD, RME and GTL is provided in Section S8.

In a collaborative effort, personal samples were also taken at the sampling site to assess the resultant exposure to mine workers during this sampling campaign [27]. Methylated PAHs and parent PAHs were found to be more prevalent in the pure diesel combustion emissions than the RME and GTL emissions. The authors reported that the highest gas phase concentrations for the targeted aromatic species were attributed to the pure diesel followed by GTL and RME. Combustion emissions from RME and GTL released lower amounts of chemical compounds associated with adverse health effects, and the authors concluded that the substitution of petroleum-based fuels could benefit employees working in confined areas. When considering noxious emissions underground, advanced combustion due to higher octane of biodiesel and GTL, as well as the increased concentration of oxygenated species in biodiesel, typically result in slightly increased  $\text{NO}_x$  emissions resulting from the higher temperatures. This can be overcome through engine recalibration to

retard injection timing with the net effect being the same  $\text{NO}_x$  emissions as with diesel but with PM emissions reduced substantially.

The work presented here confirms an earlier study by Geldenhuys et al. which characterised PAHs in various underground locations. In this earlier study, the authors found that PAHs were predominantly present in the gas phase with naphthalene and mono-methylated naphthalene being the most abundant at concentrations ranging from  $0.01$  to  $18 \mu\text{g m}^{-3}$ . The highest concentrations of particle bound PAHs were found at the exhaust of the idling load haul dump vehicle with a dominance of fluoranthene and pyrene and a total particle associated PAH concentrations ranging from  $0.47$  to  $260 \text{ ng m}^{-3}$ . Heavier PAHs detected in the filter samples included benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and benzo[ghi]perylene [15].

#### *Changes in the physical values of functional performance indicators of the trackless mobile machinery*

The extensively established advantage of using biodiesel is the reduction in pollutant concentrations from the exhaust emissions which is why over the last few years, it has been reported that biodiesel production capacity has a double-digit annual growth rate [19,38].

Biodiesel and GTL can be used directly in diesel engines without the need for any modification and in the context of implementation in the mining industry, this is key as it will allow for the continuous operation of the current LHD fleet and prevent costly and time intensive replacement. However, the use of alternative fuels with different chemical compositions and local conditions leads to the distinctive performance of the diesel engine. Studies on the performance of a diesel engine with blends of biodiesel have reported higher brake specific fuel consumption and exhaust gas temperatures when using biodiesel but a decrease in brake thermal efficiency and the fuel energy conversion efficiency is dependent on both the engine performance conditions as well as the biodiesel inclusion percent in the diesel [20,24,39]. To decrease the time and costs associated with experimental research, computational models and new mathematical-statistical methods have been shown to be very valuable tools in predicting and optimising peak engine outputs and various engine parameters such as the study by Ashok et al., whereby multi-objective response surface methodology was used to obtain engine characteristics for different fuel blends, specifically 1-pentanol/jatropha oil blends [40].

In this study, the power and performance indicators of a diesel engine were investigated during the experimental study when a LHD was operated on pure diesel, RME and GTL as detailed in Section 2. The experimental observations of the LHD when operated with different fuels are summarised in Table 1.

The cetane number of RME is slightly higher than that of diesel but it is less volatile therefore it is characterised by higher flash point, water content and up to 25 times higher contamination therefore it may reduce the auto-ignition delay and increase the amount of fuel premixed for rapid combustion and resultantly boosts the cylinder gas temperature, creating favourable preconditions for  $\text{NO}_x$  formation. The engine performance on neat RME and its blends with diesel, as well as its emission characteristics, depends largely on the injector nozzle design, the combustion chamber, needle valve opening pressure, air-fuel mixture quality, actual start of combustion and heat release peculiarities [20]. Therefore, it must be noted that different engines may vary substantially, and the results are not always directly comparative. There are, however, commonly reported advantages of biodiesel over diesel which are summarised in Table 2 with the reported disadvantages, most of which can be mitigated with internal measures and exhaust after-treatment techniques as well as inclusion of alcohol blends [8–10,24,33].

#### *Environmental and socio-economic factors*

Sustainability is multifaceted and it recognises inherent relations

**Table 1**

Comparative qualitative observations of LHD engine performance parameters when operated on neat diesel, GTL and RME fuels.

	Diesel	GTL	RME
<b>Engine performance</b>	Status quo – no observed change.	No change at low engine speeds. Reduction in power at higher engine speeds.	No change in power was observed.
<b>Smoke opacity and volume</b>	A lot of black smoke was observed with characteristic pungent diesel fume odour.	Decreased amount of smoke and increased transparency of smoke. Slight odour.	Decreased amount of smoke and increased transparency of smoke. Slight odour.
<b>Particulate matter and soot</b>	Substantial PM and black smoke affecting visibility.	Less observed PM and soot.	Less observed PM and soot.

**Table 2**

Advantages and disadvantages of the use of biodiesel over diesel (adapted from Alalwan et al [10]).

Advantages of using biodiesel	Disadvantages of using biodiesel
Generates fewer regulated pollutant emissions of CO <sub>x</sub> , SO <sub>2</sub> , PM and HC compared to diesel.	Combustion generates higher NO and NO <sub>2</sub> than diesel (can be mitigated).
Non-toxic, non-flammable and it reduces harmful exhaust emissions, smoke and odour.	Higher cloud and pour point may cause fuel freezing and more difficulty starting the engine in cold conditions.
Higher flashpoint and high combustion efficiency with lower aromatic and sulfur content.	Degradation of lubricant will lead to shorter engine life with biodiesel.
It is safer to handle and less toxic than diesel fuel.	Power and volumetric fuel consumption is reduced with biodiesel.
The fuel is biodegradable, portable, and renewable and can be produced locally.	Dependent on the amount of waste (feedstock) available and refining is required. Biodiesel distillation is preferable to improve quality.
Does not require drilling, transportation or refinement therefore production is easier and faster than diesel.	Leads to increased oil dilution by fuel causing accelerated oil degradation (due to lower volatility of biodiesel).
Higher cost efficiency than diesel.	Lack of aromatics causes fuel system seal failure unless standard nitrile seals are replaced with Viton seals.
Lower PM emissions may result in a potential reduction of required underground airflow and thereby a reduction in associated ventilation costs.	Resistance to change.

between environmental, economic and social well-being as shown in Table 3 where the current use of diesel is compared to the prospective use of greener alternatives, BTL and biodiesel. These fuels offer more sustainable solutions that can be realised relatively quickly with the current infrastructure and expertise in SA. To add to this, research by Brent et al reviewed the short-term viability of the SA biofuels strategy with respect to the environmental, economic and social conditions of sustainability and the uncertainties pertaining to the three pillars that are identified in the National Biofuels Task Team feasibility study [41].

A sector-based approach could ease the rollout of biofuel penetration: as an example, two major industries in South Africa could collaborate for mutual benefit. SA is among the world leading producers of high-quality sugar with an approximate average production of 2.3 million tons per season [42]. Pre-harvest sugar cane burning is common practice in SA where over 90 % of the sugar cane is burnt to improve harvest, handling and milling efficiencies [43]. These burn events result in atmospheric pollution, including emissions of semi-volatile organic compounds [44], that have adverse impacts on air quality and human health on a local, regional and even a global scale. An alternative to pre-harvest burning is “green harvesting” which involves the cutting of the adult cane stalk and the removal of leaves and unwanted matter which is not typically done as it increases harvest time and costs. The increase in cost for labour could be offset by remuneration for the sugarcane trash as biomass feedstock for BTL fuel. Not only will this have positive socio-economic impacts by additional job creation in the sugarcane industry, it will also result in the reduction of biomass burn emissions and lead to healthier more sustainable development in the sugar industry,

whilst at the same time providing a sustainable feedstock for alternative fuel in the mining industry strongly supporting sustainable development.

Another promising avenue that would promote a circular economy would be a collaborative effort with the forestry industry and synthetic fuel production industry, which would close the loop between feedstock producers, refining industry and fuel user. The mining industry is currently a large off taker of poor-quality timber for mine support beams used underground so integration with a forestry company would not only be feasible but also beneficial as waste foliage is potentially a good second-generation waste source. In this sense, the major sectors of the country can lead by example in finding greener solutions and striving for carbon neutrality and pave the way for the establishment of a biofuel industry.

#### Uncertainty in PAH measurements

Sources of uncertainty can arise from the sampling of analytes as well as from the analytical measurement thereof which can affect the accuracy of the experimental data.

During sampling, sources of uncertainty were minimised by the inclusion of field and instrument sampler blanks, the taking of duplicate samples and processing and handling of each sampler in the exact same manner. The sampling pumps were calibrated to minimise uncertainties regarding sample volumes. Upwind samples were taken to account for any background concentrations of analytes and this was subtracted from the exhaust samples. Direct thermal desorption of samples prior to analysis prevented any sample preparation sourced uncertainty or contamination from solvent use.

Sources of analytical measurement uncertainty were minimised by employing best practice, such as performing calibration in duplicate, using isotope-labelled internal standards and tuning the instrument each morning. The sampling and analytical methods were based on published methods [15,44,45].

#### Recommendations for future work

Due to the nature of the sampling campaign whereby a LHD was removed from the working fleet and dedicated to experimental measurements which took place in a productive operational underground mining environment, certain limitations were presented. Restricted time, space and resources during the campaign limited the number of experiments that could be conducted, and the challenging underground environment further limited the number and type of equipment that could be used for testing.

In order further evaluate the reduction of unregulated emissions when transitioning to biofuel and add to the results reported here on the PAH concentrations, it is recommended that a more comprehensive study be done which includes a larger sample set and the parallel monitoring of regulated pollutants. It is further recommended that the desorption of heavier particulate phase PAHs from QFFs be optimised, and the characterization and quantification of emissions of toxic oxygenated and nitrated PAH derivatives be investigated.

**Table 3**

Comparison of environmental, economic and social impacts pertaining to the use of alternate fuels. The plus symbols indicate the extent of the positive impact and the minus symbols represent the negative impact.

	Diesel	BTL	Biodiesel
<b>Environmental Impact</b>		++ Reduction of harmful atmospheric emissions. Reduction in GHG emissions. Sustainable feedstock. Low indirect land-use change impact. Positive environmental impact if sugarcane trash used as there will be a reduction in pre-harvest burn emissions.	+++ Reduction of harmful atmospheric emissions. Reduction in GHG emissions. Sustainable feedstock for 2 <sup>nd</sup> generation biodiesel. Positive environmental impact if sugarcane trash used as there will be a reduction in pre-harvest burn emissions. Land-use change impact. Saving of fossil fuels.
	-- Unstable fossil fuel dependence. Diesel exhaust emissions toxic to environment.		
<b>Economic Impact</b>	+ Technology and methods are established, and no additional expenditure required.	+++ Energy sufficiency and self-sufficiency. Refining power available in SA. Infrastructure: filling stations and routes of distribution can continue to be used. Fuels can be tailored to the needs of different types of engines. Farmers can benefit by selling biomass for refining. Job creation will contribute to SA GDP. CAPEX for required de-centralised plants. Increased fuel cost. Increased production cost and additional R&D required.	+++ Energy sufficiency and self-sufficiency. Refining power available in SA. Independence of energy imports and rising oil prices. Infrastructure: filling stations and routes of distribution can continue to be used. Fuels can be tailored to the needs of different types of engines. Farmers can benefit by selling biomass for refining. Job creation will contribute to SA GDP. Increased fuel cost. Increased production cost and additional R&D required.
	-- Oil price dependant. Prices likely to continuously increase due to unsustainable reserves. Taxes and penalties may be implemented in future.	- Cost of implementation in terms of infrastructure, crop cultivation logistics, processing, and exportation. Given the low volume production of biofuels they are often linked to crude-oil derived fuels in price and	- Cost of implementation in terms of infrastructure, crop cultivation logistics, processing, and exportation.

**Table 3 (continued)**

	Diesel	BTL	Biodiesel
		offer little saving to the end user unless the end user backward integrates into biofuel production.	
<b>Social Impacts</b>		+++ Strengthening of regional agriculture. Job creation. A range of different raw materials can be used. No competition for land with food production.	++ Strengthening of regional agriculture. Job creation. A range of different raw materials can be used. No competition for land with food production.
	-- Adverse health impacts from carcinogenic diesel exhaust emissions.		

**Conclusion**

Environmental protection and the search for cleaner, more sustainable fuels are two of the most important concerns modern society is facing. The projected negative impacts on climate change and human health as well as the volatility of oil supply are forcing governments to search for alternative options. The mining sector is one of the key industries contributing to the South African economy, but at the same time is one of the leading energy intensive sectors that needs more sustainable solutions. The present work focuses on the feasibility of a transition to alternative greener fuels with focus on biodiesel and biomass-to-liquid fuel in the platinum mining industry of South Africa. This study aimed to fill gaps in the literature by examining the effects of two different biofuels, namely GTL and RME used at 100% in a heavy duty LHD engine under real-world underground operation, on unregulated PAH emissions which have adverse effects on the environment, human health and can cause engine failure. The sampling of PAHs in both gas and particulate phase was achieved using small, portable denuder devices that are practical for challenging environments such as in underground mines.

Total target PAH emissions from a high-idling load haul dump vehicle were found to decrease considerably when substituting diesel fuel with gas-to-liquid or biodiesel with total PAH concentrations of 34; 14 and 9 µg m<sup>-3</sup> for emissions from the diesel, GTL and RME fuelled engine respectively. The PAH emissions resulting from both biofuels were found to be lighter 2-4 ringed PAHs in the gas phase, therefore the sampling of only PM, which is most commonly reported, would result in a gross underestimation of PAH emissions. Overall, it is concluded that use of both GTL and RME results in a considerable decrease in total PAH emissions during high idle mode and during the test cycle mode when compared to diesel.

The evaluation of the PAH emissions, although promising, cannot be used as a stand-alone indicator for the feasibility of alternative fuels but must be used in conjunction with the consideration of the environmental and socio-economic factors that South Africa faces upon implementation. These selected test biofuels have the potential of being generated locally with sustainable crops for first or second generation of biofuel, such as from sugar cane, sunflower oil or waste biomass, and can be produced in large quantities without affecting food supplies. Considering all factors, biomass residues are an increasingly attractive resource for sustainable bioenergy production in SA as not only can it help achieve a circular economy in SA, but it will also have numerous

environmental and socio-economic benefits for the country.

#### Funding sources

Funding provided by the University of Pretoria, Impala Platinum Ltd. and the National Research Foundation of South Africa (NRF, grant number 105877) is acknowledged.

#### CRediT authorship contribution statement

**G. Geldenhuys:** Methodology, Validation, Formal analysis, Data curation, Writing – original draft, Writing – review & editing. **M. Watrus:** Methodology, Writing – review & editing. **M. Fox:** Funding acquisition, Writing – review & editing. **P.B.C. Forbes:** Conceptualization, Visualization, Project administration, Methodology, Formal analysis, Writing – review & editing, Funding acquisition.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

#### Acknowledgements

The Department of Chemistry at the University of Pretoria as well as Impala Platinum Ltd are acknowledged for their support and resources. LECO is acknowledged for the use of ChromaTOF Tile software.

#### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ref.2023.100500>.

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