Diesel particulate matter and polynuclear aromatic hydrocarbon exposure of diesel vehicle operators in small to medium sized underground coal mines

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PREFACE

Occupational Hygienists responsible for underground mines are faced with numerous challenges in providing a safe working environment for employees. To provide unpolluted fresh air for breathing purposes is only one of the functions of ventilation. Other functions of ventilation are to remove heat, particles and gasses suspended in the air. The wide use of mobile diesel vehicles in underground intake airways makes it more difficult for the Occupational Hygienist to provide clean air for mine employees. Personal exposure to diesel exhaust particles and gasses needs to be established and proper controls need to be implemented to lower personal exposure. Very little exposure information is currently available for the small to medium sized coal mines in South Africa for diesel particulate matter and polynuclear aromatic hydrocarbons. These two entities are also not fully regulated in South Africa.

The main objective of this study is to determine the diesel particulate matter and polynuclear aromatic hydrocarbon exposure of operators working on LHD vehicles in small to medium sized coal mines in South Africa. This study examines international exposure limits and the lack of South African exposure limits. This study also describes the monitoring basics applicable for diesel particulate matter monitoring in coal mines.

It was decided to use the article format for this mini-dissertation. Thus, for sake of uniformity, the whole mini-dissertation is done according to the guidelines of the chosen magazine for possible publication, i.e. Annals of Occupational Hygiene. This magazine requires that the references be set out in Vancouver style.

AUTHORS AND CONTRIBUTORS

Table 1 provides the names of the authors and their contribution in the planning and execution of the study.

Table 1: List of authors and their respective contributions

Name	Contribution
Mr JM Pretorius Principle researcher	Researching relevant literature, negotiating participation, conducting monitoring, interpreting results and writing mini-dissertation.
Prof. F.C. Eloff: Supervisor	Assisting with the design and planning of the study, reviewing literature, reviewing results and advising on the interpretation of the results.
Mr P.J. Laubsher: Co-supervisor	Assisting with the design and planning of the study, reviewing literature, reviewing results and advising on the interpretation of the results.

The declaration below confirms each author's contribution in this study.

I hereby declare that I have approved the article and that my contribution in the study, as indicated above, is representative of my actual contribution. I hereby give my consent that it may be published as part of Mr JM Pretoriu's M.Sc (Occupational Hygiene) mini-dissertation.

_____ _____

Prof. F.C. Eloff **Mr P.J. Laubsher**

Supervisor **Co-supervisor**

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LIST OF ABBREVIATIONS

ACGIH	American Conference of Government Industrial Hygienists
ATSDR	Agency for Toxic Substances and Disease Registry
BOM	Bureau of Mines
CCME	Canadian Council of Ministers of the Environment
DMR	Department of Mineral Resources
DoL	Department of Labour
DPM	Diesel particulate matter
EC	Elemental Carbon
IARC	International Agency for Research on Cancer
LHD	Load Haul Dump
MHSA	Mine Health and Safety Act, Act 29 of 1996, South Africa
MSHA	Mine Safety and Health Administration
NIOSH	National Institute for Occupational Safety and Health
OC	Organic Carbon
OEL	Occupational Exposure Limit
OSHA	Occupational Safety and Health Administration
PAH	Polycyclic aromatic hydrocarbon, or Polynuclear aromatic hydrocarbon
PEL	Permissible Exposure Limit
ROS	Reactive Oxygen Species
SANAS	South African National Accreditation System
SIMRAC	The Safety in Mines Research Advisory Committee
TC	Total Carbon
USEPA	United States Environmental Protection Agency

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ABSTRACT

Limited data is currently available for diesel particulate matter (DPM) and polynuclear aromatic hydrocarbon (PAH) exposure in underground coal mines in South Africa. The lack of regulatory exposure limits for DPM and PAH in South Africa makes it difficult for the mining industry to evaluate concerned exposure results effectively. The purpose of this study was to determine load haul dump (LHD) vehicle operator exposure to DPM and PAH in four small to medium sized coal mines. Exposures were measured against international standards which could appropriately be implemented in South Africa. The National Institute for Occupational Safety and Health (NIOSH) method 5040 was used to measure exposure for DPM using submicron elemental carbon as surrogate. NIOSH method 5515 was used to measure exposure towards seventeen PAH compounds. Exposure results for DPM were far below the available exposure limit. The use of two LHD vehicles underground showed results that were 2.6 times higher than when one LHD vehicle was used. Exposure results for PAH showed values below laboratory detection limits. The TC/EC values for the four coal mines indicated that the Mine Safety and Health Administration (MSHA) TC/EC value of 1.3 is not suitable for South African coal mines if the MSHA standards for DPM were to be adopted in South Africa. The findings of this study are consistent with another local study conducted in 2008. To conclude, this study emphasized the urgency to establish regulatory exposure limits for DPM and PAH.

OPSOMMING

Baie min informasie is huidiglik beskikbaar vir diesel partikel materie (DPM) en polinukluêre aromatiese koolwaterstowwe (PAH) blootstelling in ondergrondse steenkool myne in Suid Afrika. Die gebrek aan blootstellings drempels vir DPM en PAH in Suid Afrikaanse wetgewing maak dit vir die mynbou industrie moeilik om blootstellingsresultate effektiewelik te vergelyk. Die doel van die studie was om die DPM en PAH blootstelling van laaivoertuig (LHD) operateurs in vier klein tot middelslag grote steenkoolmyne te bepaal. Blootstellingsresultate is vergelyk met internasionale blootstellings standaarde wat ook in Suid Afrika ge-implimenteer kan word. Die 'National Institute for Occupational Safety and Health' (NIOSH) metode nommer 5040 is gebruik om te DPM blootstelling in terme van sub-mikron grote elementêre koolstof te meet. NIOSH metode nommer 5515 is gebruik om PAH blootstelling vir sewentien veskillende PAH verbindings te meet. Blootstellingsresultate vir DPM was vêr onder die beskikbare blootstellingsdrempel gemeet. DPM blootstelling was 2.6 keer hoër gewees wanneer daar twee LHD voertuie saam ondergrond gewerk het in vergelyking met wanneer net een LHD voertuig alleen gewerk het. Blootstellingsresultate vir PAH was laer as die laboratorium se deteksie vlakke. Die TC/EC waardes van die vier steenkoolmyne het baie van die verwysings waarde van 1.3, soos gestel deur die 'Mine Safety and Health Administration' (MSHA), verskil. Suid Afrika sal nie die MSHA standaarde vir DPM kan implimenteer nie omdat die MSHA TC/EC waarde nie verteenwoordigend is vir Suid Afrikaanse steenkoolmyn kondisies nie. Die bevindinge van die studie was ook ooreenstemmend met 'n ander soortgelyke studie wat in 2008 lokaal uitgevoer was. Ten slotte het die studie die noodsaaklikheid beklemtoon om DPM en PAH standaarde in wetgewing te stel.

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CHAPTER 1: GENERAL INTRODUCTION

1.1 Background

The use of Load Haul Dump (LHD) diesel vehicles in underground mines is a cause of concern for occupational hygiene practitioners in the mining industry. Diesel LHD vehicles are primarily used to load, haul and dump ore and ore residue underground. Diesel exhaust particles present in underground atmospheres may pose a high risk towards the health and safety of employees working in these environments. Due to its submicron size, diesel particles remain suspended in the air for a prolonged period of time and are therefore, also not easily removed from the general air. According to Stanton *et al*. (2007), diesel exhaust emissions contain a complex mixture of gasses, vapours and particulates notably carbon monoxide, carbon dioxide, sulphur dioxide, nitrous oxides, polynuclear aromatic hydrocarbons (PAH's) and carbonaceous particles. Currently no occupational exposure limits have been assigned to diesel particulate matter (DPM) in terms of elemental carbon and most polynuclear aromatic hydrocarbon (PAH) compounds by the Department of Mineral Resources (DMR) in South Africa.

For DPM the main route of entry into the human body is by inhalation. PAH can enter the body via inhalation, the digestive system and absorption through the skin. Exposure of humans to DPM has been associated with acute inflammatory response in the peripheral blood and airway tissues as well as the increased potential for allergic reactions (Van Niekerk *et al.*, 2002). The International Agency for Research on Cancer (IARC) classified a PAH compound like benzo[a]pyrene as a human carcinogen (IARC, 1989). The IARC classified dibenzo[a,h]anthracene as a probable human carcinogen and benzo[a]anthracene, benzo[k]fluoranthene, chrysene, indeno[1,2,3-cd]pyrene and naphthalene as possible human carcinogens (IARC, 1989). Diesel exhaust as a whole was classified as a suspected occupational carcinogen by the National Institute for Occupational Safety and Health in 1988 (Van Niekerk *et al.*, 2002). The IARC classified diesel exhaust as 'probably carcinogenic in humans' in group 2A (IARC, 1989).

An important physical characteristic of diesel particles is that they are very small in size. Over 80 per cent of diesel exhaust particles are between 0.1 and 0.2 µm in diameter (Van Niekerk *et al.*, 2002). This means that it would penetrate the deepest part of the lung, the alveoli, where oxygen enters the bloodstream. Other toxins, like PAHs, can easily attach or absorb into DPM due to its

rough solid nature (Stanton *et al*., 2007). The combined exposure to DPM and PAH may pose an increased risk to the health of employees. LHD operators could most probably be exposed to larger amounts of DPM and PAH than other employees. Personal exposure of employees and diesel machinery operators to DPM and PAH matter needs to be determined in order to ensure a safe working environment. This study will provide information for DPM and PAH exposure in small to medium sized coal mines in South Africa and may also indicate the need to establish Occupational Exposure Limits (OEL's) in South African regulations.

1.2 Problem statement

Limited data are available for personal exposure to DPM and PAH in coal mines in South Africa. Currently no accepted occupational exposure limits (OEL's) have been assigned to DPM in coal mines for South Africa. There is no DPM OEL for coal mines set in the United States of America except for a tailpipe standard of 2.5 g/h (Stanton *et al*., 2007). The National Institute for Occupational Safety and Health (NIOSH) regulates two PAH compounds, the Occupational Safety and Health (OSHA) regulates five PAH compounds and the American Conference of Government Industrial Hygienists (ACGIH) regulates one PAH compound (NIOSH, 1994). In South Africa only one PAH compound is regulated. This study will focus on the DPM and PAH exposures of diesel LHD operators working underground in small to medium sized coal mines.

1.3 Objectives of the study

The main objective of this study is to quantify LHD operator's exposure to DPM and PAH in four small to medium sized coal mines in South Africa. For the purpose of this study small is seen as < 250 employees and medium is seen as 250 to 500 employees. This study will also describe the concerned markers and monitoring equipment that are applicable to determine accurate personal exposure. This study may indicate the need for OEL's to be set for DPM and PAH in South Africa and comment on the adoption of international standards.

1.4 Hypothesis

LHD operators working underground in small to medium sized coal mines are exposed to levels of DPM and PAH that exceed available standards.

1.5 Research design

Four small to medium sized coal mines participated in the study. Four LHD operators, one on each coal mine, were identified to participate in the study. Two samples, one for DPM and one for PAH, were taken on the day shift for a period of five consecutive days at each mine. Each LHD operator wore two sampling pumps, one for sampling DPM and one for sampling PAH, for an 8 hour period. The inlets of both sampling cassettes were placed next to each other on the left shoulder within a 30cm sphere from the nose and mouth of the employee. NIOSH method 5040 was used for determining DPM exposure and NIOSH method 5515 was used to determine PAH exposure. Filter cassettes and sorbent tubes were sent to a SANAS accredited laboratory for analysis. Results were compared with standards if available.

1.6 References

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CHATER 2: LITERATURE REVIEW

2.1 Physical and chemical characteristics of DPM and PAH

Diesel contains a mixture of hydrocarbons or compounds containing hydrogen and carbon atoms. When diesel is used as a fuel in the burning and combustion process in engines to propel vehicles, certain by-products are formed by the combustion process and from the evaporation of diesel itself (Van Niekerk *et al.*, 2002). These by-products are commonly referred to as diesel exhaust. According to Stanton *et al*. (2007), diesel exhaust emissions contain a complex mixture of gasses, vapours and particulates notably carbon monoxide, carbon dioxide, sulphur dioxide, nitrous oxides, polynuclear aromatic hydrocarbons (PAHs) such as benzo[a]pyrene, aldehydes, unburnt hydrocarbons and carbonaceous particles.

Diesel particulate matter (DPM) has a carbonaceous core with absorbed organic compounds and small amounts of sulfates, metals and other compounds. The carbonaceous core is defined as elemental carbon (EC) and the absorbed organic compound is defined as organic carbon (OC) (Vermeulen *et al*., 2010). DPM is defined as a sub-micron (< 1.0 micron) physical aerosol component of diesel exhaust (DPM is made up of solid carbon particles, organic chemicals and inorganic compounds). The carbon component, known as total carbon (TC), is made up of EC and OC. TC makes up about 85 % of DPM (Belle, 2008).

According to the Canadian Council of Ministers of the Environment (CCME, 2010), PAHs are a group of complex hydrocarbons comprised of two or more fused benzenoid rings. PAHs, also known as polycyclic aromatic hydrocarbons, are a group of chemicals that are formed during the incomplete burning of coal, oil, gas, wood or other organic substances such as tobacco. There are more than 100 different PAHs. PAHs occur as complex mixtures and not as single compounds. As pure chemicals PAHs exist as colorless, white or pale yellow-green substance with a faint pleasant odour. PAHs are used in the manufacturing of dyes, plastics, pesticides and are also found in substances such as oil, coal, coal tar pitch, creosote and roofing tar. PAHs may be found in air, water and soil. PAHs enter environments mostly as releases into the air from volcanoes, forest fires, wood burning and exhaust from automobiles and trucks. PAHs in the air can travel long distances before returning to the earth in rainfall or particle settling. PAHs can also evaporate from soil and water containing PAH compounds. PAHs can break down to longer-lasting products by reacting with

sunlight and other chemicals in the air, usually over a period of days to weeks. Some PAH compounds can also attach to dust particles present in the air (ATSDR, 1995). PAHs are often divided into low molecular weight PAHs and high molecular weight PAHs. Low molecular weight PAHs tend to have a core structure of two to three benzenoid rings. High molecular weight PAHs tend to have molecular structures of four or more benzenoid rings. The hydrophobicity, tendency for bioaccumulation, resistance to biodegradation, and overall environmental persistence of PAHs generally increase with increasing molecular weight. Low molecular weight PAHs are more water soluble than high molecular weight PAHs. Low molecular weight PAHs tend to be more volatile than high molecular weight PAHs. High molecular weight PAHs also tend to be more lipophilic than low molecular weight PAHs (CCME, 2010).

2.2 Route of entry into human body for DPM and PAH

Over 80% of diesel exhaust particles are between 0.1 and 0.2 µm in diameter and remain suspended in the air for long periods of time. DPM penetrate deep into the lung, via inhalation, and locate across the respiratory epithelium (Van Niekerk *et al.*, 2002). Particles may deposit by five major mechanisms i.e. inertial impaction, gravitational sedimentation, Brownian diffusion, electrostatic attraction and interception (USEPA, 2002). Inertial impaction results when particles collide with the airway walls due to a change in air stream direction. The particle weight and the flow rate of the air stream causes the particle to keep its trajectory, away from the changing air stream, resulting in impaction against the airway walls. Inertial impaction is applicable for particle diameters > 5µm and becomes an important deposition mechanism further down the respiratory system for particles < 2 µm. Gravitational sedimentation refers to the settlement of particles due to gravity. Gravitational sedimentation becomes the dominant mechanism of deposition for particles ranging from 0.5 µm to 5 µm in diameter. Deposition by Brownian diffusion occurs when particles collide with gas molecules resulting in a random motion. Deposition by Brownian diffusion increases with decreasing particle size and becomes the dominant mechanism for deposition for particles > 0.05 µm in diameter. Deposition by electrostatic precipitation results from charged particles inducing image charges of opposite sign onto the surfaces of the airways that are electrically conducting while normally uncharged. Charged particles become electrostatically attracted to airway walls, and the consequence is that charged particles deposit on airway walls. Deposition by electrostatic precipitation is less than 10% of total deposition. The interception mechanism occurs in the absence

of the previous mechanisms when the particle dimension is the same as the airway dimension, resulting in deposition (Darquenne, 2006). The relative contribution of each deposition mechanism to the fraction of inhaled particles deposited varies for each region of the respiratory tract (USEPA, 2002).

PAH compounds may enter the human body via inhalation in the lungs when air containing PAH is inhaled. Sources of PAHs include cigarette smoke, smoke from burning coal and wood, industrial waste sites, diesel exhaust, cooking meat on open fires and dust containing PAH compounds. PAHs can enter the body via the gastrointestinal tract if taken in orally. PAH absorption in the human body is slow when drinking water containing PAHs, swallowing dust and eating food containing PAH. Under normal conditions of environmental exposure PAHs can enter the human body through the skin when in contact with soil containing high levels of PAH, products like creosote and crank case oil. Lipophilic PAHs, benzo[a]pyrene can be absorbed or even be stored in the stratum corneum of the skin. These compounds may enter all tissues in the body containing fat tissue. Primarily these compounds will be stored in the liver, kidneys and other fat tissue. Smaller amounts will be stored in the spleen, adrenal glands and the ovaries. PAH compounds are also changed from their initial state into other states by the tissue in the body. When a change in the compound's state occurs, it may be more or less harmful to the body compared to its original state (ATSDR, 1995).

2.3 Factors influencing exposure to DPM and PAH

Control parameters available to lower exposure to DPM must be utilized. Underground mining controls include the use of low sulphur fuel, good diesel engine maintenance programmes, exhaust after treatment, proper ventilation control, control of the number of diesel vehicles emitting DPM and unnecessary idling of diesel vehicles (Belle, 2008). The use of old diesel engine technology compared to more technological advanced diesel engines may contribute to lower DPM levels.

Human exposure to PAHs is expected to be highest among certain occupational groups like individuals working with coal tar, foundry workers, miners and chimney sweeps. Smokers themselves and people living with them or in their vicinity are exposed to PAHs. People consuming smoked or grilled food, fish and shellfish or people making use of heating appliances burning wood or coal are more susceptible to PAH exposure (ATSDR, 1995).

The susceptibility of people within populations exposed to PAHs may differ with age, genetic makeup, dietary habits, health and nutritional status and substance exposure history. These parameters result in decreased function of the detoxification and excretory processes (mainly hepatic, renal, and respiratory) or the pre-existing compromised function of target organs including effects or clearance rates and any resulting end-product metabolites. Children with developing organs and elderly people with declining organ function are more vulnerable to toxic substances than adults (ATSDR, 1995).

2.4 Health effects and mechanisms

2.4.1 Diesel particulate matter

2.4.1.1 Symptoms

Sydbom *et al.* (2001) point out that acute health effects after exposure to DPM entails irritation of the nose and eye, lung function changes, airway inflammation, headache, fatigue and nausea. According to Sydbom *et al.* (2001), the chronic health effects of exposure to DPM entails sputum production, coughing and lung function decrements. There were also several observations supporting the hypothesis that diesel exhaust fumes are a contributing factor towards the allergy pandemic. Whether DPM is introduced intraperitoneally, intranasally or intratracheally it may act as adjuvant to allergen and thus increase the sensitization process. In addition to that, exposure studies in healthy humans have documented profound inflammatory changes in the airways before changes in pulmonary functions are detected. These effects might be more detrimental in humans with asthma and humans with compromised pulmonary function.

2.4.1.2 Effects on humans

Bayram *et al*. (2006) conducted a study describing the regulation of human lung epithelial cell numbers during diesel exhaust particle exposure. Bayram *et al*. (2006) indicated that under a normal situation of an intact epithelium, in the absence of serum with no inflammatory response, the epithelial cells are under a balanced turnover of proliferation and apoptotic cells. The data indicated that at low levels of exposure diesel exhaust particles may induce hyperplasia of a normal epithelium by preventing cell apoptosis. In the presence of inflammation with serum extravasation,

the effects of diesel exhaust particles on cell numbers are masked by those of serum. Serum itself induces proliferation. Oxidative stress factors may underlie the diesel exhaust particle-induced induction of apoptosis of epithelial cells. Antioxidants can protect against diesel particle-induced increase in epithelial cells.

Baulig *et al*. (2003) concluded that diesel exhaust particles, via their organic components, modify the cellular redox state. They provided evidence that organic compounds are bioavailable as they induce gene expression and can be metabolized. Finally diesel exhaust particles and their extracts induce, in human bronchial and nasal epithelial cells, the expression of numerous genes as well as in the secretion of proinfammatory cytokines (Baulig *et al*., 2003).

Li *et al*. (2002) demonstrate that organic diesel exhaust particle extracts, including polar and aromatic fractions, induce oxidative stress in epithelial cells, in response to which these cells exhibit induction of apoptosis-necrosis. Epithelial cells produced more super oxide radicals and were more susceptible to cytotoxic effects than macrophages. Cytotoxicity is the result of mitochondrial damage, ROS production and ATP depletion. If of sufficient intensity, oxidative stress can initiate proinflammatory effects in macrophages and bronchial epithelial cells. These effects are mediated by phosphorylation-dependent cell signaling pathways, including activation of the mitogen-activated protein kinase cascades. Another consequence of oxidative stress is the induction of cellular apoptosis and necrosis. Diesel exhaust particles induce cellular apoptosis and necrosis change of the mitochondrial PT pore. This leads to a cascade of events that include cytochrome C release and activation of cellular caspases. Damage to the mitochondrial inner membrane also disrupts four-electron reductions of O_2, switching this process instead to one-electron reduction. Epithelial cells are also more susceptible to cytotoxic effects of diesel exhaust particle extracts than macrophages.

Li and Nel (2006) explained that airway epithelial cells are at great risk of DNA damage when constantly exposed to particulate matter-induced oxidative stress and carcinogens. In the presence of DNA damage, a control mechanism that induces cell cycle arrest is activated to ensure the fidelity of DNA replication and genomic integrity.

2.4.1.3 Effects on animals

Hougaard *et al*. (2008) conducted a study to determine whether diesel exhaust exposure in mice would affect gestation, postnatal development, activity, learning or memory and biomarkers of transplacental toxicity. Findings indicated that gestational parameters were similar between control and diesel exhaust exposed groups. The difference increased during lactation. Diesel exhaust particle exposed offspring weighed significantly less than the control progeny. Slight effects of exposure were observed on cognitive function in female diesel exhaust particle exposed offspring and on biomarkers of exposure to particles or genotoxic substances.

Long-term studies for diesel exhaust exposure in rats demonstrated increased accumulation of particles and aggregates of particle laden macrophages in the alveoli and peribronchial interstitial tissues as well as local inflammation, epithelial proliferation, fibrosis and emphysematous lesions. In a study exposing rats to increasing concentrations of diesel exhaust particles for up to 24 months, a progressive increase in the lung burden of particles was seen at the highest concentrations of diesel exhaust particles (Sydbom *et al*., 2001).

When mice were exposed to high concentrations of diesel exhaust particles, an intratracheal instillation of diesel exhaust particles was found to cause severe lung injury and high mortality. The cause of death was pulmonary oedema mediated by endothelial cell damage. The toxicological effect and increased mortality were to a great extent prevented by pretreatment with the oxygen radical scavenger superoxide dismutase, supporting the hypothesis that diesel exhaust particle toxicity is connected to production of radical superoxide O_2^{-1} leading to endothelial cell damage. Pathological and histological studies in rats found an increase in lung weight, increased number of particles in the lung associated with alveolar infiltration of macrophages, macrophage aggregation, chronic inflammatory responses, proliferation and hyperplasia of alveolar epithelium and type 2 cells, thickening of alveolar septa and wall fibrosis (Sydbom *et al.*, 2001).

2.4.2 Polynuclear aromatic hydrocarbons

2.4.2.1 Symptoms

Acute effects associated with PAH exposure, which include headache, nausea, respiratory and dermal irritation, are probably caused by other associated agents. Overall PAHs have a low acute

toxicity. Naphthalene, the most abundant constituent of coal tar, is a skin irritant and its vapours may cause headache, nausea, vomiting and diaphoresis (ATSDR, 2009).

2.4.2.2 Effects on humans

Effects reported as a result of chronic occupational exposure to PAHs include chronic bronchitis, chronic cough irritation, bronchogenic cancer, dermatitis, cutaneous photosensitization and pilosebaceous reactions. Reported health effects associated with chronic exposure to coal tar pitch and its byproducts include irritation and photosensitivity in the eyes. Chronic exposure effects for the skin include erythema, burns, warts on sun-exposed areas with progression of cancer. The toxic effects of coal tar on the skin are enhanced by exposure to ultraviolet light. Coughing, bronchitis and bronchogenic cancer in the respiratory system are signs of chronic exposure to PAHs. Leukoplakia, buccal-pharyngeal cancer in the lip is associated with chronic exposure to PAHs. Leukemia, lymphoma, hematuria and bladder and kidney cancers are associated with chronic exposure to PAHs (ATSDR, 2009). Excess incidences of lung cancer have been associated with PAH exposure in a variety of occupational settings, including coal gassification, coke production, paving and roofing, and various occupations involving exposure to creosote or soot (CCME, 2010). There is evidence of an increased risk for skin and scrotal cancers from occupational exposure to creosote and coal tar, while excess incidences of stomach and colorectal cancer have been observed for coal gas production workers (CCME, 2010).

Benzo[a]pyrene (BaP) is readily absorbed following inhalation, ingestion and skin exposure. Following inhalation and ingestion, BaP is rapidly distributed to several tissues in rats, including the kidney, small intestine, trachea, stomach, testes, liver and oesophagus. BaP is metabolized by cytochrome P450 enzymes resulting in a number of metabolites being formed, including the reactive epoxide metabolite, BaP 7,8 diol-9,10-epoxide, which is believed to be responsible for its carcinogenicity (HPA,2008).

Cavallo *et al*. (2005) showed oxidative DNA damage due to exposure to asphalt fumes in paving workers chronically exposed to low doses of PAH mixtures. Long-term low-level exposure to PAHs and halogenated aromatic hydrocarbons has been associated with a wide variety of effects including irritability, mood instability, short and long-term memory loss, and lack of concentration in children (Dahlgren *et al*., 2003).

Table 2 provides the carcinogenic classification for some PAH compounds according to the IARC monographs on the evaluation of carcinogenic risks to humans, volume 92 (IARC, 2010). Table 3 represents the IARC classification system of carcinogenicity for humans (IARC, 2010).

Table 2: IARC carcinogen classification of PAH compounds.

Polynuclear aromatic hydrocarbons	Carcinogen classification
Benzo[a]pyrene	1
Dibenzo[a,h]anthracene	2A
Benzo[a]anthracene	2B
Benzo[k]fluoranthene	2B
Chrysene	2B
Indeno[1,2,3-cd]pyrene	2B
Naphthalene	2B
Acenaphthene	3
Anthracene	3
Benzo[b]fluoranthene	3
Benzo[ghi]perylene	3
Benzo[e]pyrene	3
Fluoranthene	3
Phenanthrene	3
Pyrene	3

Table 3: The IARC classification for carcinogenicity.

Group 1	The agent is carcinogenic to humans
Group 2A	The agent is probably carcinogenic to humans
Group 2B	The agent is possibly carcinogenic to humans
Group 3	The agent is not classifiable as to its carcinogenicity to humans
Group 4	The agent is probably not carcinogenic to humans

Benzo[a]pyrene is classified as a human carcinogen. Dibenzo[a,h]anthracene is classified as a probable human carcinogen and benzo[a]anthracene, benzo[k]fluoranthene, chrysene, indeno[1,2,3-

cd]pyrene and naphthalene are classified as possible human carcinogens. Insufficient evidence is available to define the combined effect of DPM and PAH exposure in humans or animals. The PAH and other organic compounds attaching onto the particulate fraction of diesel exhaust gasses may result in a possible additive effect, but this will remain uncertain until more scientific evidence becomes available.

2.4.2.3 Effects on animals

PAHs like benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-c,d]pyrene caused tumur growth in laboratory animals when animals were exposed via inhalation, skin contact and gastro intestinal intakes. When pregnant mice were fed benzo[a]pyrene they had difficulty reproducing. Their offspring also have difficulty reproducing. The offspring have also shown teratogenic effects and decreased body weight. Animal studies have also shown that exposure to PAH compounds had harmful effects on the immunologic system of the body to combat disease when exposed over the short-term or the long-term (ATSDR, 1995).

BaP can cross the placenta and was found to cause adverse developmental and reproductive effects in mice. Dietary administration during gestation reduced fertility and fetal abnormalities whereas administration by gavage caused an increase in fetal death and decreased fertility. Short-term dietary administration of BaP caused forestomach tumours in mice and hamsters. Chronic exposure of mice to BaP by gavage or in the diet resulted in forestomach and lung tumours and in rats an increase in tumours of the forestomach, oesophagus, liver, larynx and mammary gland was observed. Chronic inhalation of BaP caused an increase in lung tumours in mice, and tumours of the nasal cavity, pharynx, trachea, oesophagus and forestomach in hamsters (HPA, 2008).

2.5 Protection against exposure

The first step to prevent exposure to any hazardous substance would be to eliminate the substance from occurring or substituting the substance with another substance not affecting health (COSHH, 2008). Should the first step not be practical, the second step would be to control exposure by:

- Total enclosure of the source, if possible.
- Limiting the area to which exposure occurs.

- Use of local exhaust systems.
- Dilution of hazardous substance by means of ventilation.
- Reducing the period of exposure.
- Providing suitable approved personal protective equipment as a last resort (COSHH, 2008).

Humans exposed to acute high dosages of PAHs should remove contaminated clothing and scrub the skin gently with soap and water. Chronic exposed humans should undergo periodic medicals to identify adverse health effects and to initiate treatment (ATSDR, 2009).

2.6 Measurement Methodology

2.6.1 Diesel particulate matter

The NIOSH method 5040 is commonly accepted to be the appropriate method for determining personal exposure to diesel particulate matter in terms of EC (Standton *et al.*, 2007). This method targets the particulate carbon portion of diesel exhaust. In the analysis the TC on a filter is quantified by the summation of OC and EC. In the absence of interferences TC was the logical surrogate to determine exposure to DPM, because DPM is mostly >80% carbon. Unfortunately other OC sources made TC measurements interference prone. This made the determination of EC to be the better surrogate for occupational exposure to DPM (Birch and Noll, 2004). Following this EC monitoring strategy according to NIOSH 5040, provided acceptable results for EC to determine DPM exposure in non-coal mines. In coal mines the EC interference from coal dust would make it difficult to determine DPM exposure accurately. Most diesel particles have aerodynamic diameters of less than 1µm and most coal dust particles have aerodynamic diameters of bigger than 1µm. This means that a size selective sampling strategy could be followed to separate DPM from coal dust. This led to the development of the Bureau of Mines (BOM) sampler development whereby 90%, based on particle mass, of coal dust was removed. Based on the BOM impactor design, the SCK® DPM cassette was developed and tested successfully to become an alternative for monitoring DPM in coal mines (Noll & Birch, 2004). For this study NIOSH method 5040 in conjunction with the SCK® DPM cassette were used to determine DPM exposure in small to medium sized coal mines.

2.6.2 Polynuclear aromatic hydrocarbon

NIOSH method 5515 was used to determine exposure to specific PAH compounds in this study. Although NIOSH method 5506 is more sensitive than NIOSH method 5515, the laboratory used for analysis was only accredited for NIOSH method 5515. NIOSH method 5515 provided information on the vapour and particle fractions of the PAH compounds tested for. Special precautions were taken to reduce interference from heat, ozone, NO_2 and Ultraviolet light.

2.7 Occupational exposure limits

2.7.1 Diesel particulate matter

For metal mines in the USA the current MSHA Permissible Exposure Limit (PEL) for DPM is 350 µg/m³ TC and 270 µg/m³ EC (Belle, 2008). The TC/EC ratio used was 1.3. Belle (2008) investigated the TC/EC ratios in different South African mines. Up to date there is no OEL standard for EC determination of DPM for coal mines in the USA. The only standard existing is a motor vehicle tail pipe standard of 2.5 g/h (Stanton *et al.*, 2007).

The Australian mining industry found with extensive research that when employees were exposed to below 0.2 mg/m³ as submicron DPM, 0.16 mg/m³ submicron TC or approximately 0.1 mg/m³ submicron EC, the level of eye and respiratory tract irritation was significantly reduced. On this basis the New South Wales Minerals Council then proposed a best practice exposure standard of 0.2 mg/m³ as submicron DPM, 0.16 mg/m³ submicron TC and 0.1 mg/m³ submicron EC. The Minerals Council also acknowledged that although there was substantial improvement in employees comfort by using this standard, there was not sufficient evidence suggesting that this standard would prevent the development of cancer (Stanton *et al.*, 2007).

Currently in South Africa no exposure limit is prescribed by the Department of Mineral Resources (DMR) or the Department of Labour (DoL) for DPM in terms of EC or TC.

2.7.2 Polycyclic aromatic hydrocarbon

Table 4 shows the exposure limits set by different organisations. This information was sourced from NIOSH 5515 (1994).

Table 4: Polynuclear aromatic hydrocarbon exposure limits.

	OSHA	NIOSH	ACGIH
acenaphthene	-	-	-
acenaphthylene	-	-	-
anthracene	0.2 mg/m³	-	-
benz[a]anthracene	-	-	-
benzo[b]fluoranthene	-	-	-
benzo[k]fluoranthene	-	-	-
benzo[ghi]perylene	-	-	-
benzo[a]pyrene	0.2 mg/m³	0.1 mg/m³	-
benzo[e]pyrene	-	-	-
chrysene	0.2 mg/m³	-	-
dibenz[a,h]anthracene	-	-	-
fluoranthene	-	-	-
fluorine	-	-	-
indeno[1,2,3-cd]pyrene	-	-	-
naphthalene	10 ppm	10 ppm	10 ppm
phenanthrene	0.2 mg/m³	-	-
pyrene	-	-	-

It is clear that seventeen different compounds can be analyzed but not enough information is available to define the negative effect each compound poses to the health of employees. Of the seventeen compounds OSHA regulates five compounds, NIOSH regulates two compounds and ACGIH regulates one compound. It is clear that a large amount of research still needs to be done on PAH compounds to determine the health effects exposure will have on employees. This information could then be used to verify regulatory standards to be set. OSHA has set a Permissible Exposure Limit (PEL) for PAHs as a group of 0.2 mg/m³ in terms of coal tar pitch volatiles in 1977. OSHA also promulgated a PEL of 0.15 mg/m³ for coke oven emissions in 1978 (Van Niekerk *et al.*, 2002).

In South Africa only Naphthalene is regulated by the DMR with a occupational exposure limit of 10 ppm.

2.8 Related studies

In June 2002 SIMRAC published a study to determine diesel particulate emissions in the South African mining industry (Van Niekerk *et al.*, 2002). Two coal mines and one gold mine were selected to participate in this study. The primary objective of this study was to identify the most appropriate sampling and analytical methods currently available to measure diesel particulate emissions. DPM and PAH were measured in the three mines. Results indicated high exposures to DPM in these mines. This study also indicated that no standard was available for coal mines in terms of DPM. Results indicated that exposure to PAHs was found to be insignificant. The study highlighted that NIOSH method 5040 in terms of elemental carbon was still the preferred method for monitoring personal exposure and that the same method together with size selective sampling could be used to determine elemental carbon in coal mines. In this study the need to set exposure standards for DPM in South Africa was pointed out.

Birch and Noll (2004) indicated that a standard for DPM based on particulate carbon was not considered practical in coal mines due to the interference of coal dust. The study also indicated that interference from coal dust may not be a problem should an appropriate size selective sampler be used and an appropriate elemental carbon (EC) standard be set. This study showed that EC contributed by sub micrometer coal dust was minor when size selective sampling was done. This study also pointed out the total carbon (TC) and EC differences between different types of coal. These differences will have a huge impact on the establishment of a standard in terms of EC for coal mines.

Belle (2008) investigated the TC/EC ratios in different South African mines to determine whether the same TC/EC ratio, used by the MSHA, could be applied to set a EC standard based on a TC standard. In the United States of America the MSHA proposed a standard for metal mines of 350 µg/m³ for TC and 270 µg/m³ for EC with a TC/EC ratio of 1.3. The TC/EC ratio plays an important role as EC is seen as a specific sensitive marker for DPM. This study indicated a mean TC/EC ratio of 1.44 ranging from 1.25 to 2.13. In this study results from DPM monitoring done on different

South African mines indicated significant differences in the TC/EC ratios from one mine to the next. Significant differences were also observed when comparing South African TC/EC ratios with other countries. Therefore, it is accepted that the TC/EC ratio will be different from mine to mine and even from section to section.

Coble *et al*. (2010) conducted air monitoring surveys at seven non-metal mining facilities between 1998 and 2001. Exposure to Respirable elemental carbon (REC) was assessed for an epidemiological study of miners exposed to diesel exhaust fumes. Personal exposure measurements were taken on workers in different job categories on the surface and underground. REC measurements were used to develop quantitative estimates of average exposure levels by facility, department and job title for epidemiologic analysis. The average REC, smaller than 10 µm, exposure level for underground jobs with five or more measurements ranged from 31 µg/m³ to 58 µg/m³ at the facility with the lowest exposure levels. Average REC values at the facility with the highest exposure ranged from 313 µg/m³ to 488 µg/m³. The average exposure for surface workers ranged from 2 µg/m³ to 6 µg/m³ across the seven facilities. In total 80% of underground jobs were assigned exposure levels based on measurements that were taken.

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CHAPTER 3: ARTICLE

Diesel particulate matter and polynuclear aromatic hydrocarbon exposure of diesel vehicle operators in small to medium sized underground coal mines

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Keywords: Diesel particulate matter, Polynuclear aromatic hydrocarbon, Total carbon, Elemental Carbon, Occupational exposure limit, NIOSH 5040, NIOSH 5515, Coal mines.

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• Where individuals need to be specified for certain sources of funding the following text should be added after the relevant agency or grant number 'to [author initials]'.

An example is given here: 'This work was supported by the National Institutes of Health [AA123456 to C.S., BB765432 to M.H.]; and the Alcohol & Education Research Council [hfygr667789].'

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3.2 Abstract

Limited data is currently available for diesel particulate matter (DPM) and polynuclear aromatic hydrocarbon (PAH) exposure in underground coal mines in South Africa. The lack of regulatory exposure limits for DPM and PAH in South Africa makes it difficult for the mining industry to evaluate concerned exposure results effectively. The purpose of this study was to determine load haul dump (LHD) vehicle operator exposure to DPM and PAH in four small to medium sized coal mines. Exposures were measured against international standards which could appropriately be implemented in South Africa. The National Institute for Occupational Safety and Health (NIOSH) method 5040 was used to measure exposure for DPM using submicron elemental carbon as surrogate. NIOSH method 5515 was used to measure exposure towards seventeen PAH compounds. Exposure results for DPM were far below the available exposure limit. The use of two LHD vehicles underground showed results that were 2.6 times higher than when one LHD vehicle was used. Exposure results for PAH showed values below laboratory detection limits. The TC/EC ratios for the four coal mines indicated that the Mine Safety and Health Administration (MSHA) TC/EC ratio of 1.3 is not suitable for South African coal mines if the MSHA standards for DPM were to be adopted in South Africa. The findings of this study are consistent with another local study conducted in 2008. To conclude, this article emphasized the urgency to establish regulatory exposure limits for DPM and PAH.

3.3 Introduction

The use of mobile diesel machinery in underground environments is a cause of concern for management structures in terms of the health and safety of employees working underground. Limited information is currently available in terms of exposure of employees towards diesel exhaust emissions in the small to medium sized coal mines in South Africa.

According to Stanton *et al.* (2007), diesel exhaust emissions contain a complex mixture of gasses, vapours and particulates notably carbon monoxide, carbon dioxide, sulphur dioxide, nitrous oxides, polynuclear aromatic hydrocarbons (PAH), aldehydes, unburnt hydrocarbons and carbonaceous particles or soot. Diesel particulate matter (DPM) is also not easily removed from the general air due to its sub micron size and thus remains suspended in the air for a prolonged period of time (Belle, 2008). Organic compounds like PAH can easily be absorbed into DPM where they are then transported to affected areas in the human body (Stanton *et al*, 2007). According to Van Niekerk *et al*. (2002), human exposure to DPM has been associated with increased numbers of alveolar macrophages, T-lymphocytes and neutrophils in the broncho-alveolar lavage fluid, as well as acute inflammatory response in the peripheral blood and airway tissues and increased potential for allergic reactions. A PAH compound like benzo[a]pyrene is classified as a human carcinogen by the International Agency for Research on Cancer (IARC, 2010). Diesel exhaust as a whole was classified as a suspected occupational carcinogen by the National Institute for Occupational Safety and Health in 1988 (Van Niekerk *et al.*, 2002). The objective of this study is to provide information in terms of the exposure of mobile diesel machine operators towards DPM and PAH in small to medium sized coal mines in South Africa.

The coal mining industry, locally and internationally, is faced with certain problems in setting occupational exposure standards (OEL) for DPM. Currently there is no OEL set for DPM exposure in South Africa. The problems pertaining to setting an OEL in coal mines make it difficult to adopt an international OEL for DPM exposure, should such an OEL exist. In the United States of America there is still no accepted OEL for DPM exposure in coal mines, accept for a tailpipe standard of 2.5 g/h (Stanton *et al.*, 2007). From the seventeen PAH compounds investigated according to the National Institute for Occupational Safety and Health (NIOSH) method 5515, South Africa only set an OEL for one compound. The Occupational Safety and Health Administration (OSHA) has listed five compounds for which OEL have been set. It is clear that more research has to be done as to

understand the adverse health effects of different compounds on human health. This information must be used to set safe OEL's against which human exposures could be measured.

3.4 Occupational Exposure Limits

Historically the concerned marker used for DPM was TC. TC is made up of the sum of EC and OC. The reason TC was used as a surrogate to measure DPM exposure is that it represented about 80% of diesel exhaust. As professionals were using TC as the concerned marker they found that various organic substances originating from minerals dusts, burning oil and cigarette smoke interfered with achieving accurate TC results for DPM exposure. Most interfering substances were organic substances. The OC component of TC was then removed resulting in EC becoming the logical surrogate to determine DPM exposure (Belle, 2008). The focus was then to model an EC standard from the TC standard. The mining industry was interested in finding a conversion factor between TC and EC. The establishment of such a conversion factor would grant access to previous data and it would allow comparison with results from other field studies where gravimetric analysis was not possible (Noll *et al*., 2007). The TC/EC ratio was then used as the conversion factor. The TC/EC ratio basically defines the role of OC interference.

Currently in South Africa (SA) no exposure limit is prescribed by the Department of Mineral Resources (DMR) or the Department of Labour (DoL) for DPM in terms of EC or TC. Non-coal mines may refer to international standards but coal mines cannot. Worldwide there is no accepted standard specifically, for DPM in terms of EC, for coal mines.

In 2002 the ACGIH proposed a standard of 0.02 mg/m³ but this proposal was withdrawn in 2003. In 2008 the MHSA in the USA was phasing in a Permissible Exposure Limit (PEL) for DPM of 350 µg/m³ for TC and 270 µg/m³ for EC for non-coal mines (Stanton *et al.*, 2007). The TC/EC ratio used was 1.3 (Stanton *et al.*, 2007).

For SA to adopt this standard as set by the MSHA, the TC/EC ratio of 1.3 would have to be representative for SA mines. Belle (2008) investigated the TC/EC ratios in different SA mines and found a mean TC/EC ratio of 1.44, ranging from 1.25 to 2.13. According to these results, Belle (2008) indicated that care should be taken in the adoption of international standards.

Up to date there is no OEL standard for EC determination of DPM for coal mines in the USA. The only standard existing is a tail pipe standard of 2.5 g/h (Stanton *et al.*, 2007).

The Australian mining industry found with extensive research that when employees were exposed to below 0.2 mg/m³ as submicron DPM, 0.16 mg/m³ submicron TC or approximately 0.1 mg/m³ submicron EC, the level of eye and respiratory tract irritation was significantly reduced. On this basis the New South Wales Minerals Council then proposed a best practice exposure standard of 0.2 mg/m³ as submicron DPM, 0.16 mg/m³ submicron TC and 0.1 mg/m³ submicron EC (Stanton *et al.*, 2007). The Minerals Council acknowledged that although there was substantial improvement in employees comfort by using this standard, there was not sufficient evidence suggesting that this standard would prevent the development of cancer (Stanton *et al.*, 2007).

To measure DPM in coal mines a size selective sampling strategy is followed to prevent interference from coal dust EC (Noll & Birch, 2004). In coal mines EC smaller than 1 µm (submicron) is measured to determine exposure to DPM (Noll & Birch, 2004). The standard of 0.1 mg/m³ submicron EC as proposed by the New South Wales Council will be used for this study due to particle size similarity and non-interference from coal particle EC above 1 µm.

For PAHs the following exposure limits as shown in Table 1 exist. Information in Table 1 below was sourced from NIOSH 5515, 2003.

Table 1: Polynuclear aromatic hydrocarbon exposure limits.

	OSHA	NIOSH	ACGIH
acenaphthene	-	-	-
acenaphthylene	-	-	-
anthracene	0.2 mg/m³	-	-
benz[a]anthracene	-	-	-
benzo[b]fluoranthene	-	-	-
benzo[k]fluoranthene	-	-	-
benzo[ghi]perylene	-	-	-
benzo[a]pyrene	0.2 mg/m³	0.1 mg/m³	-
benzo[e]pyrene	-	-	-
chrysene	0.2 mg/m³	-	-
dibenz[a,h]anthracene	-	-	-
fluoranthene	-	-	-
fluorine	-	-	-
indeno[1,2,3-cd]pyrene	-	-	-
naphthalene	10 ppm	10 ppm	10 ppm
phenanthrene	0.2 mg/m³	-	-
pyrene	-	-	-

It is clear that seventeen different compounds can be analyzed, but not enough information is available to define the negative effect each compound poses to the health of employees. Of the seventeen compounds OSHA regulates five compounds, NIOSH regulates two compounds and ACGIH regulates one compound. It is clear that a large amount of research still needs to be done on PAH compounds to determine the health effects exposure will have on employees. This information could then be used to verify regulatory standards to be set. OSHA have set a Permissible Exposure Limit (PEL) for PAHs as a group of 0.2 mg/m³ in terms of coal tar pitch volatiles in 1977 (Van Niekerk *et al.*, 2002). OSHA also promulgated a PEL of 0.15 mg/m³ for coke oven emissions in 1978 (Van Niekerk *et al.*, 2002). In South Africa only naphthalene is regulated by the DMR with a occupational exposure limit of 10 ppm.

3.5 Methods and Instrumentation

Four small to medium sized coal mines were selected and agreed to participate in the study. One mobile diesel machine operator and machine were selected to participate in the monitoring process. An operator was issued with two Gillian pumps, connected to a sampling train for DPM and another sampling train for PAH. Both monitoring cassettes were placed in the operators breathing zone. The inlets of both cassettes were placed next to each other on the left-hand side of the operator's chest. Monitoring was conducted over an eight hour day shift for five consecutive days at each coal mine. The operators conducted their normal day to day work underground.

NIOSH method 5040 was used to determine DPM exposure in terms of elemental carbon. A calibrated Gillian air sampling pump was connected to the SKC® DPM cassette with integral precision-jeweled impactor and quartz-fiber filters were used for monitoring DPM exposure at these coal mines. Diesel particulate matter is characterized as small particles smaller than 1.0 µm (Stanton *et al.*, 2007). Mechanized coal dust suspended in the air, normally bigger than 1.0 µm, is excluded by the GS-1 cyclone and the impactor present in the SKC® DPM cassette. The GS-1 cyclone with a flow rate of 1.7 L/min. rejects dust particles bigger than 4.0 µm, 50 % cutoff point, to prevent clogging and obstruction in the impactor present in the SKC® DPM cassette. The impactor in the SKC® DPM cassette allows any particles smaller than 1.0 µm through to be collected on the first filter. The design of this size selective sampling system is based on the Bureau of Mines (BOM) impactor which removed 90% of the coal dust interference (Noll & Birch, 2004). Particles collected on the first filter are a combination of elemental carbon and organic carbon. The SKC® DPM cassette is, therefore, equipped with a second filter installed in tandem from the first filter in the same cassette. According to Noll and Birch (2004), the purpose, in theory, of the second filter is to absorb organic carbon smaller than 1 µm which, after analysis, is to be subtracted from the first filter containing both elemental carbon and organic carbon to provide a corrected elemental carbon value. Samples were analyzed according to the NIOSH method 5040 and results were obtained in terms of elemental carbon, organic carbon and total carbon.

NIOSH method 5515 was used to determine the following PAH compounds:

acenaphthene	benzo[ghi]perylene	fluorine
acenaphthylene	benzo[a]pyrene	indeno[1,2,3-cd]pyrene
anthracene	benzo[e]pyrene	naphthalene
benz[a]anthracene	chrysene	phenanthrene
benzo[b]fluoranthene	dibenz[a,h]anthracene	pyrene
benzo[k]fluoranthene	fluoranthene	

A Gillian sampling pump was connected to a XAD-2 sorbent tube and filter cassette containing a 37 mm quartz-fiber filter in tandem were used. The Gillian pump was calibrated to a flow rate of 2 L/min. The filter collected the airborne particles and the sorbent tube collected the vapours. Both the filter and the sorbent tube were analyzed using NIOSH method 5515. All analyses were done by an accredited laboratory.

3.6 Statistical Analyses

Results from the laboratory were corrected to an 8 hour Time Weighted Average value to make comparison with OEL and PEL limits possible. A five day mean was then calculated to assign an exposure value for the five day monitoring period per coal mine. Where applicable the standard deviation was utilized to determine how widespread day to day results were from the 5 day mean values in the different coal mines. The 5 day mean value per coal mine was then compared in terms of percentage of the applicable OEL or PEL.

Where laboratory results indicated exposures below the individual compounds detection limit the following procedure was implemented:

a) The data presented was treated utilizing the formulae $D/\sqrt{2}$ (Ogden, 2010).
b) Then an 8 hour TWA correction was made to accommodate differences in sampling volumes.
c) These treated results were then used to determine a five day average for each coal mine.

3.7 Results

3.7.1 DPM Results

Table 2 shows the five day monitoring results for EC at the different coal mines. Large variations in EC monitoring results between the different days are noted per small to medium sized coal mine. Coal mine A shows much higher EC values compared to the other coal mines. The mean (M) EC exposures of all mines are much lower than the exposure limit (EL).

Table 2: EC results for the different coal mines.

Elemental Carbon	Day 1 (µq/m³)	Day 2 (µq/m³)	Day 3 (µq/m³)	Day 4 (µq/m³)	Day 5 (µq/m³)	Mean (µq/m³)	STDEV (µq/m³)	EL (µq/m³)	M % of EL
Mine A	1.68	0.66	0.08	1.38	2.02	1.16	0.79	**100**	1.16
Mine B	0.53	1.07	0.26	0.26	0.01	0.43	0.40	**100**	0.43
Mine C	0.20	0.09	0.55	0.73	0.23	0.36	0.27	**100**	0.36
Mine D	0.15	0.60	0.55	0.21	0.27	0.36	0.21	**100**	0.36

Table 3 shows the five day monitoring results for TC at the different coal mines. Similar to EC results large TC monitoring results variations are noted between the different days per mine. Coal mine A shows much higher TC values compared to the other coal mines. The mean EC exposures of all mines are much lower than the exposure limit.

Table 3: TC results for the different coal mines.

Total Carbon	Day 1 (µq/m³)	Day 2 (µq/m³)	Day 3 (µq/m³)	Day 4 (µq/m³)	Day 5 (µq/m³)	Mean (µq/m³)	STDEV (µq/m³)	EL (µq/m³)	M % of EL
Mine A	1.83	0.74	0.13	1.48	2.15	1.27	0.82	**160**	0.79
Mine B	0.75	1.18	0.52	0.52	0.03	0.60	0.42	**160**	0.38
Mine C	0.31	0.13	0.73	1.01	0.56	0.55	0.35	**160**	0.34
Mine D	0.26	0.76	0.78	0.40	0.45	0.53	0.23	**160**	0.33

Figure 1 shows the five day mean EC and TC results for the different coal mines. Coal mine A shows much higher mean exposure values than coal mines B, C and D. Coal mines B, C and D shows similar results. The difference between coal mine A and the other coal mines is most probably due to more than one mobile diesel machine being operational in the same area. Overall the results show very low exposures when compared to available standards.

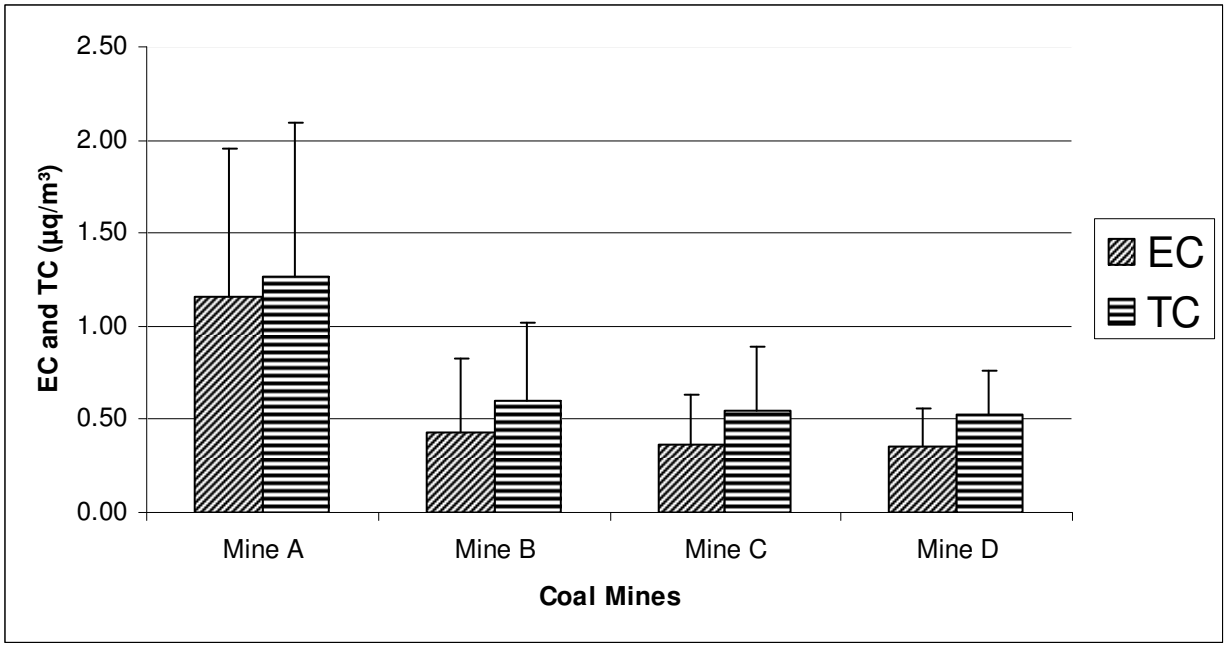


Figure 1: Mean EC and TC exposure for different mines.

Table 4 shows the calculated TC/EC ratios of each day for the different coal mines. Significant differences are seen between different monitoring days per mine. The mean TC/EC ratios of the different mines are significantly different from the reference ratio of 1.3.

Table 4: TC/EC ratios for the different coal mines.

TC/EC Ratio	Day 1	Day 2	Day 3	Day 4	Day 5	Mean TC/EC	STDEV	TC/EC Ref	M % of Ref	% Diff From 1.3
Mine A	1.09	1.12	1.63	1.07	1.06	1.19	0.24	**1.3**	91.88	-8.12
Mine B	1.42	1.10	2.00	2.00	3.00	1.90	0.72	**1.3**	146.43	46.43
Mine C	1.55	1.44	1.33	1.38	2.43	1.63	0.46	**1.3**	125.23	25.23
Mine D	1.73	1.27	1.42	1.90	1.67	1.60	0.25	**1.3**	122.92	22.92

The TC/EC ratios for the different coal mines are presented in Figure 2. There is a difference in the TC/EC ratios between the different coal mines. The TC/EC ratio for coal mine A is lower than the reference ratio of 1.3 and the TC/EC ratios for coal mine B, C and D are higher than the reference ratio.

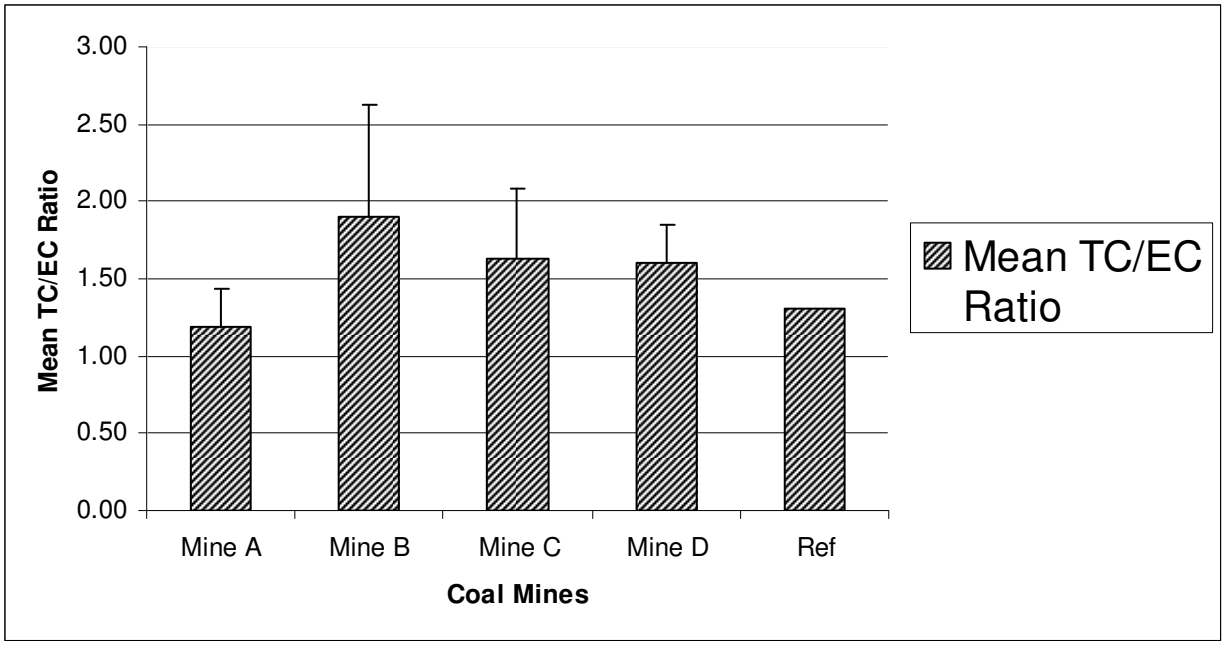


Figure 2: Mean TC/EC ratios for different mines.

Figure 3 shows the percentage difference of each of the different coal mines TC/EC ratios from the reference TC/EC ratio of 1.3. Large differences can be seen above and below the reference TC/EC ratio of 1.3.

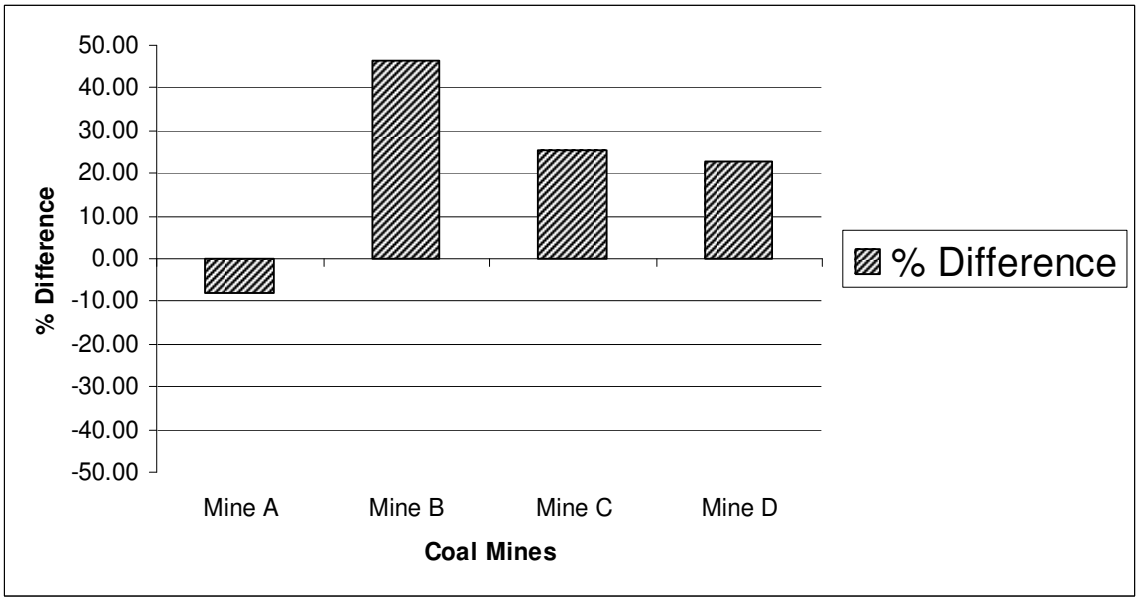


Figure 3: % difference from TC/EC Ratio of 1.3.

3.7.2 PAH Results

Table 5 provides the five day mean PAH results for the different coal mines. Below detection limit results from the laboratory were treated according to the procedure explained in section 3.6 above to provide corrected results as shown in Table 9. Overall results show very low exposure levels.

Table 5: PAH 8 hour Time Weighted Average monitoring results.

PAH Compound (µg/m³)	OSHA OEL	Mine A (5 Day Mean)	Mine B (5 Day Mean)	Mine C (5 Day Mean)	Mine D (5 Day Mean)
ACENAPHTHENE		0.101	0.141	0.141	0.141
ACENAPHTHYLENE		0.056	0.080	0.082	0.080
ANTHRACENE	**200 µg/m³**	0.070	0.100	0.301	0.100
BENZ[A]ANTHRACENE		0.174	0.245	0.249	0.253
BENZO[B]FLUORANTHENE		0.115	0.161	0.162	0.164
BENZO[K]FLUORANTHENE		0.448	0.642	0.639	0.641
BENZO[GHI]PERYLENE		0.244	0.357	0.349	0.353
BENZO[A]PYRENE	**200 µg/m³**	0.244	0.357	0.349	0.353
BENZO[E]PYRENE		0.115	0.161	0.162	0.164
CHRYSENE	**200 µg/m³**	0.143	0.201	0.602	0.204
DIBENZ[A,H]ANTHRACENE		0.244	0.357	0.349	0.353
FLUORANTHENE		0.084	0.121	0.122	0.120
FLUORENE		0.070	0.100	0.101	0.100
INDENO[1,2,3-CD]PYRENE		0.240	0.357	0.349	0.353
NAPHTHALENE (**ppm**)	**10 ppm**	0.00001	0.00002	0.00002	0.00002
PHENANTHRENE	**200 µg/m³**	0.070	0.100	0.101	0.100
PYRENE		0.101	0.141	0.141	0.144

3.8 Discussion

3.8.1 Diesel particulate matter

Table 2 provides the five day monitoring results for EC for coal mines A, B, C and D. The standard deviations for each coal mine indicate that there were substantial differences between the day to day EC exposures. The mean EC exposure for coal mine A was 1.16 µg/m³ with day to day EC exposures ranging from 0.08 µg/m³ to 2.02 µg/m³. The mean EC exposure for coal mine B was 0.43 µg/m³ with a range of 0.01 µg/m³ to 1.07 µg/m³. Coal mine C shows a mean EC exposure of 0.36 µg/m³ ranging from 0.09 µg/m³ to 0.73 µg/m³. The mean EC exposure for coal mine D was also 0.36 µg/m³ with a range of 0.15 µg/m³ to 0.60 µg/m³. The same tendency and variations can also be seen in the TC results shown in Table 3. The variations in the ranges and standard deviation values indicate that exposure monitoring conditions were not the same between the different days on a mine and even between different mines. These variations can be mainly attributed to different working patterns. The working patterns of these LHD vehicles for all four coal mines are not the same and are also different on each mine on a day to day basis. Activities range from transporting material in and out of the coal mines to different areas underground, traveling different routes underground with different ventilation conditions and sometimes working together with other LHD vehicles or alone.

EC and TC results given in Table 2 and Table 3 indicate that coal mine A has a substantially higher mean exposure than the rest of the coal mines. This higher mean exposure for coal mine A is illustrated in Figure 1 where the mean TC and EC exposures are given per coal mine. Only one LHD vehicle was operational underground at coal mines B, C and D. Two LHD vehicles were operational underground in coal mine A. The applicable LHD vehicle in coal mine A worked together with another LHD vehicle in the same area when monitoring was conducted. The LHD operator at coal mine A is exposed to higher concentrations of EC due to the presence of another LHD vehicle in the same working area. The mean EC values for each coal mine are very low when compared against the exposure limit (EL) of 100 µg/m³. The mean EC exposure of coal mine A is the highest of the four coal mines at only 1.16% of the EL. The mean EC exposure for coal mine A is also 2.6 times higher than the highest EC mean of the other coal mines. The EC exposure at coal mine A has more than doubled with double the amount of LHD vehicles that were operational underground. For this study these low EC exposures could be attributed to LHD vehicles working and traveling in

sufficiently ventilated open underground areas and not working the full 8 hours on which monitoring was conducted.

Should South Africa look towards adopting a PEL of 350 µg/m³ for TC and 270 µg/m³ for EC with a TC/EC ratio of 1.3 as set by the MHSA, the TC/EC mean value for South African mines should be very close to 1.3. Figure 2 provides the mean TC/EC ratios as measured in the four coal mines. The five day mean TC/EC ratio for coal mine A was calculated at 1.19 ranging from 1.06 to 1.63. For coal mine B the mean TC/EC ratio was 1.90 ranging from 1.10 to 3.0. The mean TC/EC ratio for coal mine C was 1.63 ranging from 1.33 to 2.43. For coal mine D the mean TC/EC ratio was 1.60 ranging from 1.27 to 1.90. The mean TC/EC ratios between the different coal mines ranged from 1.19 to 1.90 with a mean TC/EC ratio of 1.58. Belle (2008) indicated TC/EC ratios for nine coal mines ranging from 1.25 to 2.13 with a mean TC/EC ratio of 1.44. Comparison of this study with the study of Belle (2008) shows a 4.8% variation outside the lower range of 1.25. The higher range TC/EC ratio of 1.90 in this study is within the higher range TC/EC ratio of 2.13 indicated in Belle's (2008) study. Apart from the range similarities between this study and the study of Belle (2008) the mean TC/EC ratio of 1.58 is 9.7% higher than the mean TC/EC ratio of 1.44 shown by Belle (2008). This significant difference, more than 5%, indicates that the TC/EC ratios from one coal mine to the next are wide spread. The large differences between the higher and lower ranges in this study and in the study of Belle (2008) indicate that it would be difficult to set a TC/EC ratio for coal mines as a whole in South Africa.

Figure 2 compares the TC/EC ratios of this study to the reference TC/EC ratio of 1.3 as set by the Mine Safety and Health Administration (MSHA) in the United States of America. Coal mine A shows a lower mean TC/EC ratio of 1.19. Coal mine B, C and D shows higher mean TC/EC ratios of 1.90, 1.63 and 1.60 respectively. Figure 3 shows the percentage difference from the reference TC/EC ratio of 1.3. Coal mine A shows a 8.12% lower ratio. Coal mine B shows a 46.4% higher mean TC/EC ratio and coal mine C shows a 25.2% higher ratio. Coal mine D shows a 22.9% higher ratio. All four coal mines mean TC/EC ratios differ with more than 8% up to 46% from the reference TC/EC ratio of 1.3. There are significant variations in mean TC/EC ratios from this study towards the reference TC/EC ratio of 1.3 as set by the MSHA.

3.8.2 Polynuclear aromatic hydrocarbons

The Time Weighted Average (TWA) results for seventeen different PAH compounds, according to NIOSH 5515, are provided for coal mines A, B, C and D in Table 5. Comparing certain PAH compounds with their respective OELs it is apparent that results of exposure are far below the OEL's, thus indicating that LHD operators, health was not seriously affected in this monitoring period. Not all PAH compounds are assigned with OELs thus making it difficult to say that these exposures are not affecting the health of LHD operators. Furthermore, incorporating the treatment of results as explained in 3.6 statistical analyses, the laboratory results indicated readings below the detection limits of each PAH compound. With current available information it is clear that overall results indicate very low exposure to PAH compounds but more research is needed to assign all PAH compounds with OELs.

3.9 Conclusion

Results for DPM exposure for LHD operators at the four small to medium sized coal mines in South Africa indicated very low exposures when compared with the submicron standard of the New South Wales Mineral Council's exposure limit of 160 µg/m³ for TC and 100 µg/m³ for EC. DPM exposures, in terms of EC, were below 1.2 % of the EC exposure limit that was used for this study. The low DPM exposures were attributed to LHD vehicles working in open, well ventilated back area conditions. Differences in day to day working patterns of LHD vehicles were detected with large variations of day to day EC exposure results and standard deviations. The average EC exposure at coal mine A was 2.6 times higher than the highest mean EC exposure of the other three coal mines. The reason for this was that coal mine A had two LHD vehicles operational underground in the same area and the other three coal mines only had one LHD vehicle operational underground.

This study also stressed the need to establish a South African coal mine exposure limit for DPM in terms of submicron EC. A submicron EC OEL will enable coal mines to compare their exposures without the interference of coal dust generated EC. Belle (2008) indicated that care should be taken to adopt international exposure limits for South African conditions. The MSHA Permissible Exposure Limit (PEL) for DPM of 350 µg/m³ for TC and 270 µg/m³ for EC with a TC/EC ratio of

1.3 could be used to regulate DPM in South Africa if the TC/EC ratio of 1.3 was representative for South African mines. Belle (2008) indicated that a TC/EC ratio of 1.3 was not representative for South African mines and that more research was needed to provide more representative TC/EC ratios for South African mines. The mean TC/EC ratios for the four coal mines in this study show similar findings. The use of the MSHA TC/EC ratio of 1.3 is not representative for the four coal mines in this study. In fact the mean TC/EC ratio determined by this study for the four coal mines also varied from the study of Belle (2008). This supports the need for more research that needs to be conducted in order to obtain more representative TC/EC ratios in coal mines in South Africa before international standards are adopted.

The standard as set by the MSHA as given above is also not health based, but is based upon the belief that it would be economically and technically feasible for mines to reach. This standard is also applicable for non-coal mines. The Australian New South Wales Minerals Council implemented a best practice exposure limit of 100 µg/m³ for submicron EC that could be used for coal mines. Although there is no scientific proof that by implementing this exposure limit, the development of cancer would be prevented, we would not be left behind the international community by using this exposure limit in South Africa. This standard was used in this study and is also being used by local mining companies in South Africa. Consideration should also be given towards the differences in sampling strategy between coal mines and non-coal mines when an exposure limit is established. For the time being mines should measure and implement the 2.5 g/h tail pipe standard for compliance. A large study covering small, medium and large mines by determining exhaust tail pipe emissions, taking into account the amount of mobile diesel vehicles used and the ventilation requirements that are needed, could lead to the establishment of a model that could be used for future mine planning purposes.

Results for PAH exposure for LHD operators at the four small to medium sized coal mines in South Africa indicated exposures below the laboratory detection limits and were very low when compared to available PELs. The overall need to establish exposure limits for all seventeen PAH compounds was stressed as well as the need for South Africa to lay down regulating exposure limits. More research towards the health effects of long-term and short-term exposures is needed to aid the process of setting exposure limits.

3.10 References

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CHAPTER 4: CONCLUDING CHAPTER

4.1 Summary

The objectives of this study have been met by successfully determining the DPM and PAH exposures of LHD operators on four small to medium sized coal mines in South Africa. When comparing DPM results from this study to the submicron exposure limit of 160 µg/m³ for submicron TC and 100 µg/m³ for submicron EC, set by the New South Wales Mineral Council, the exposure of LHD operators were very low, less than 1.2 % of the exposure limits for both TC and EC. These low exposures were attributed to sufficient ventilation being available to effective remove DPM. Coal mine A showed 2.6 times higher exposures when compared with the other three coal mines. This was due to another LHD vehicle working in the same area as the LHD vehicle that was monitored. This additive effect, is therefore, very important when planning to use any mobile diesel vehicle in underground workings.

The need to lay down exposure limits for DPM was stressed. In South Africa DPM is not regulated in terms of a personal exposure limit. There is a difference in sampling strategy between coal mines and non-coal mines in that non-coal mines do not have coal dust generated EC interference when monitoring the EC fraction in terms of NIOSH 5040. DPM in coal mines can be measured utilizing the SKC®DPM cassette with precision jeweled impactor. This method entails monitoring the submicron fraction of EC whereby 90% of coal dust EC interference is excluded. By using this method we need to lay down a regulatory submicron exposure limit for DPM in terms of submicron EC according to NIOSH 5040 in coal mines in South Africa. To lay down a regulatory personal exposure limit for DPM in terms of submicron EC is, however, not as easy as it seems. Proper research for South African conditions needs to be done to understand South African conditions compared to international conditions. Only then can such personal exposure limits be laid down and regulated. A large amount of data for DPM exposure are available from the bigger mining companies but very little data are available for the small to medium sized coal mines in South Africa. Hopefully the data presented by this study may be helpful in the assessment process to determine a submicron personal exposure limit for the South African coal mining industry.

The mean TC/EC ratios for the different coal mines indicated that the TC/EC ratio of 1.3 as implemented by the MSHA was not representative for South African coal mines. This statement is

supported by a nine coal mine study conducted by Belle (2008). In fact the mean TC/EC ratios of this study also varied from the study of Belle (2008). Both this study and the study of Belle (2008) indicated the need for more research to be done in determining a representative TC/EC ratio for South African coal mines. Except for the TC/EC ratio problem, coal mines on the other hand have an additional problem of coal dust EC interference. This problem is adequately addressed by implementing a size selective sampling strategy whereby EC, originating from diesel exhaust, is measured as particles smaller than 1 µm in size. Utilizing the SKC® DPM cassette developed from the BOM sampler, 90% of coal dust EC based on particle mass is removed. In understanding that a size selective sampling strategy can be followed to determine EC from diesel exhaust in coal mines, it is apparent that a different OEL need to be established for coal mines than for non-coal mines unless the same sampling strategy is followed. It must be stated that setting an EC standard for coal mines is not only a South African problem but also an international problem. Much more research is needed locally and abroad to set an EC OEL standard for coal mines. The Australian New South Wales Mineral Council implemented exposure limits of 0.2 mg/m³ as submicron DPM, 0.16 mg/m³ submicron TC and 0.1 mg/m³ submicron EC. Research has shown a significant reduction of eye and respiratory tract irritation when these standards were used. The New South Wales Mineral Council also acknowledged that there was no scientific evidence that the development of cancer would be prevented by using this standard. The 100 µg/m³ submicron EC standard was used in this study and is also used by local South African mining companies. South Africa would not be left behind the rest of the world by using this standard for coal mines.

In the mean time the South African mining industry should seriously implement the exhaust tail pipe standard of 2.5 g/h for underground mobile diesel vehicles. The effect of multiple LHD vehicles operational in the same underground working area was shown in this study. The use of multiple mobile diesel machinery should be taken into account when planning the ventilation requirements of an underground mine. It is recommended that more research be done on the effect of multiple mobile diesel machinery in underground environments. Such a large study should cover small, medium and large mines and take into account all different types of mobile diesel machinery used underground. This data could be used to formulate a model and classification system whereby the amount of mobile diesel machinery, ventilation requirements and tons produced per timeframe are taken into consideration. Underground mines should put more emphasis on the use of mobile diesel machinery underground and the related air quality reductions applicable. The combined additive effect between different particles, gasses and fumes was partially covered in this study. It is,

therefore, suggested that more emphasis be placed on the total air quality index, all airborne pollutants together, than just evaluating single pollutant entities individually. To conclude we should start looking at the big picture.

PAH results for five of the seventeen compounds indicate a very low risk towards exposure when measured against OSHA's OELs. The same cannot be said for the other 12 compounds for they are not regulated with OELs. In South Africa only naphthalene is regulated with an OEL. NIOSH only regulates two PAH compounds and the ACGIH only regulates one PAH compound. It is clear that more research is needed to establish the health effects of long term low concentration exposure to PAH compounds. Additional research is also needed to establish the combined health effects of PAH absorbing into EC particles.

The hypothesis, LHD operators working underground in small to medium sized coal mines are exposed to levels of DPM and PAH that exceed available standards, was not accepted for the applicable periods of measurement. Due to financial restrictions only five samples could be taken for DPM and five samples could be taken for PAH at each of the four coal mines. More samples per coal mine would definitely provide more representative results. It is, therefore, recommended that more research should be done in the small to medium sized coal mining industry to provide more representative personal exposure results for DPM and PAH.