UNIVERSITY OF THE
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JOHANNESBURG

# Modelling Risk Exposure of BTEX Emissions from a Diesel Refuelling Station in Johannesburg, South Africa

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A thesis submitted to the Faculty of Science, University of the Witwatersrand, in fulfillment of the requirements for the degree of Doctor of Philosophy.

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# DECLARATION

I declare that this thesis is my own, unaided work. It is being submitted for the degree of Doctor of Philosophy at the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination in any other University.

RMoolla.

Raeesa Moolla
Johannesburg, 04 August 2015

# ABSTRACT

Petrol and diesel fumes are known to be anthropogenic sources of air pollutants that have a negative impact on both environmental and human health. In developing countries, attendants are still employed to pump fuel for customers. In South Africa gas pump attendants refuel vehicles with various octane unleaded petrol, lead replacement petrol (LRP) and diesel on a daily basis. Attendants are particularly at risk to adverse health effects associated with inhalation of hazardous air pollutants (HAPs). Of increasing concern in recent years are the volatile organic compounds (VOCs), with particular reference to the six aromatic hydrocarbons (benzene, toluene, ethyl benzene and three isomeric xylenes), namely the BTEX.

These pollutants are known to be potentially hazardous to human and environmental health. BTEX emissions have been found to cause central nervous system depression, organ failure, teratogenic effects and gastrointestinal disorders. Other studies have argued that the release of gases can also be detrimental as they have carcinogenic agents, while other gases attack the respiratory tracts and act as asphyxiants. Regular air quality monitoring and hazard risk assessments are significant within a developing world context as relevant air monitoring information is limited.

This study analyses the concentrations and health risks of BTEX monitored at a diesel-refueling bay in Johannesburg, South Africa. Samples were obtained for the winter periods of June, July and August of 2013 and 2014. BTEX were analysed using gas chromatography (Syntech Spectras Gas Chromatography 955), coupled with a photoionization detector (PID), Radiello passive samplers and fuel data logs.

Results indicate O-xylene (29-50%) and benzene (13-33%) are the most abundant species of the total cumulative BTEX at the site. Occupational concentrations of TEX concentrations were within local and international occupational exposure (OELs) limits throughout the monitoring period, based on 8-hour time weighted averages. However, benzene concentrations were above international OELs, but below national limits, noting a large discrepancy between these limits.

The health risk assessment of BTEX found that there were incidences when BTX concentrations were above recommended standards. Furthermore, due to the poor ventilation and high exposure duration, at the study site, the average benzene concentration over the sampling campaign exceeded the US Environmental Protection Agency's chronic inhalation exposure reference concentration. Lifetime cancer risk estimation showed that on average there is a $3.78 \times 10^{-4}$ cancer risk, corresponding to an average chronic daily intake of $1.38 \times 10^{-3}$ mg/kg/day of benzene exposure. Additionally, there were incidences where individuals were at potential hazard risk of benzene and toluene that may pose non-carcinogenic effects to employees.

Additionally, it was established that $BTEX_{total}$ concentrations were positively correlated to the volume of diesel dispensed daily and inversely correlated to temperature. Ethyl-benzene and o-xylene indicated a positive correlation with volume of fuel dispensed. Proxy estimates relating to cancer and hazard risks indicate that employees are at potential risk to adverse health effects associated with inhalation exposure to these pollutants. Mitigation strategies are recommended.

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# RESEARCH OUTPUT

## PUBLICATIONS AND PRESENTATIONS:

(Presenting Author in bold)

1. **Moolla, R.**, Curtis, C.J., and Knight, J 2015: Assessment of air quality at a bus diesel-refuelling bay: A case study in Johannesburg, South Africa. *Science of the Total Environment.* (Accepted)
2. Dangor, F., Hoogendoorn, G. and **Moolla, R.**, 2015: Medical Tourism by Indian-South Africans to India: an exploratory investigation, *Bulletin of Geography: Socio-Economic Series*, 29: 19-30.
3. **Moolla, R.**, Curtis, C.J., and Knight, J. 2015: Occupational exposure of diesel station workers to BTEX compounds at a bus depot. *International Journal of Environmental Research and Public Health, 12*: 4101-4115. (DOI: 10.3390/ijerph120404101)
4. **Moolla, R.**, Curtis, C.J., and Knight, J 2014: BTEX Concentrations Influenced By External Factors At A Diesel-refuelling Station in Johannesburg, South Africa. *The Sustainable City IX, Volume2*: 1459-1467. (DOI: 10.2495/SC141232).
5. **Moolla, R.**, Berry, W.J., Curtis, C.J, and Knight, J. 2014: BTEX concentrations and associated health implications at a diesel refuelling station, in Johannesburg, South Africa. Oral paper presented at: Annual American Association of American Geographers (AAG 2015) Conference. Chicago, USA: 21-26 April 2015.
6. **Moolla, R.** and Curtis, C.J. Health Risk Assessment of BTEX emissions at a diesel station in Johannesburg, South Africa. Oral paper presented at: International Conference of Environmental and Occupational Health 2014. Putrajaya, Malaysia: 7-9 April 2014.
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Benzene and Toluene at a landfill in Johannesburg, South Africa. Oral paper presented at: the 7th international conference on Environmental Health Risk, Budapest, Hungary: 23-25 April 2013.

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13. **Moolla, R.** and Kotze, N. 2009: Quality of life and housing satisfaction in RDP houses in Braamfischerville, Soweto: A South African case study. Oral paper presented at: ix ISQOLS Conference- Quality of Life Studies: Measures and Goals of the Progress of Societies; Florence, Italy.
14. **Moolla, R.** and Kotzé, N. 2009: *Housing Satisfaction in RDP Homes in Braamfischerville, Soweto.* Poster presented at: 8th Biennial Conference of the Society of South African Geographers: Geography for a Better World; Pretoria, South Africa.
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# LIST OF ABBREVIATIONS

| | | |
|---|---|---|
| AM | : | auto-mechanic |
| AQA | : | Air Quality Act |
| AQGs | : | air quality guidelines |
| BTEX | : | benzene, toluene, ethyl-benzene and xylenes |
| $BTEX_{total}$ | : | cumulative sum of benzene, toluene, ethyl-benzene and xylenes |
| BTX | : | benzene, toluene, xylenes |
| $C_6H_6$ | : | benzene |
| $C_7H_8$ | : | toluene |
| $C_8H_{10}$ | : | xylene |
| $C_8H_{10}$ | : | ethyl-benzene |
| CNS | : | central nervous system |
| CR | : | cancer risk |
| EU | : | European Union |
| FBA | : | fuel bay attendant |
| GC | : | gas chromatography |
| HAPs | : | hazardous air pollutants |
| hPa | : | hecto Pascals |
| HR | : | hazard risk |
| HRA | : | health risk assessment |
| kPa | : | kiloPascals |
| LADD | : | lifetime average daily dose |
| m | : | meters |
| $m^3$ | : | cubic meters |
| max | : | maximum |
| mg | : | milligram |
| $mg.k^{-1}.day^{-1}$ | : | milligrams per kilogram per day |
| min | : | minimum |
| mm | : | millimetres |
| $ms^{-1}$ | : | meters per second |
| m-xylene | : | meta-xylene |
| NIOSH | : | National Institute for Occupational Safety and Health |
| OELs | : | occupational exposure limits |

| | | |
|---|---|---|
| o-xylene | : | ortho-xylene |
| PAHs | : | polycyclic aromatic hydrocarbons |
| PID | : | photo-ionisation detector |
| ppb | : | parts per billion |
| ppm | : | parts per million |
| p-xylene | : | para-xylene |
| RfC | : | reference dose concentration |
| s.d. | : | standard deviation |
| SA | : | South Africa |
| SAOHS | : | South African Occupational Health and Safety |
| SAPIA | : | South African Petroleum Industry Association |
| TEX | : | toluene, ethyl-benzene, xylenes |
| TWA | : | time weighted average |
| US EPA | : | United States Environmental Protection Agency |
| USA | : | United States of America |
| v/v | : | volume/volume |
| VOCs | : | volatile organic compounds |
| WHO | : | World Health Organization |

# CHAPTER ONE : GENERAL INTRODUCTION

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**1.3 Air quality and legislative practices in South Africa**

**1.4 Health risks associated with BTEX**

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**1.7 Study area and site description**

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**1.9 Structure of the thesis**

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## 1.1 Introduction

For the majority of the population, exposure to petrol and diesel is generally through inhalation of vapours released from petrochemical refineries, industries, vehicle emissions and at service stations during refuelling (Figure 1.1). The release of harmful air pollutants (HAPs) is through the combustion or evaporative emissions from these activities. These emissions have been studied extensively, as they could be harmful to human and environmental health, and deteriorate the air quality of the area (Bruce et al., 2002; Brunekreef and Holgate, 2002; Fenger, 1999; Han and Naeher, 2006; Rekhadevi et al., 2010; Singh et al., 2013; Sydbom et al., 2001; World Health Organization, 2012). Thus, ambient air quality guidelines have been derived, and their primary basis is to ensure the protection of human health, as air quality and health effects are intrinsically linked (Brunekreef and Holgate, 2002; Smith, 1993).

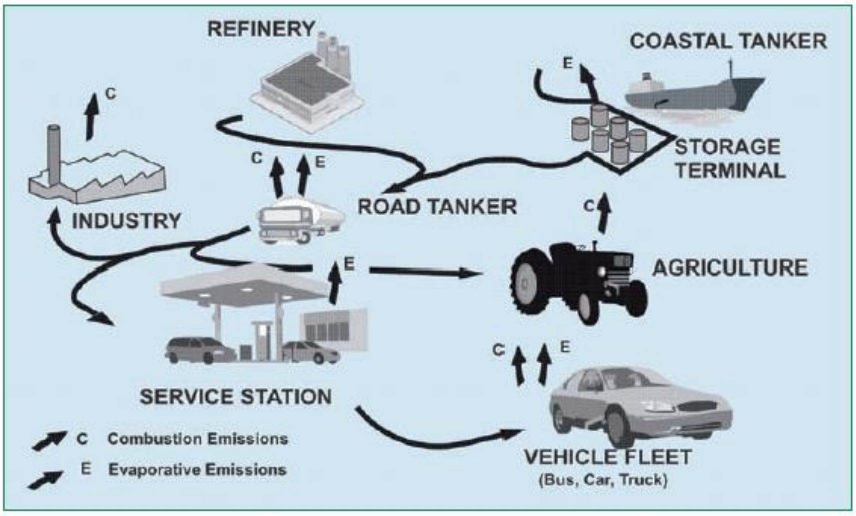

Figure 1.1: Air emissions in different environments, arising from petrol and diesel emissions, through evaporative and combustion activities (Source: SAPIA, 2008).

However, due to occupational settings, some people may be further exposed to volatile organic compounds (VOCs) that are released from diesel and petrol emissions. Of major concern are employees working in petrochemical industries, fuel tanker drivers, and forecourt attendants at service stations, as there may be increased concentrations of VOCs associated with these activities. Forecourt attendants specifically are exposed to vehicular emissions, fumes from storage vessels, as well as vapours from refuelling.

At all service stations, storage vessels will be filled with petrol and/or diesel fuel for refuelling. However, within these storage vessels, air will be displaced from the vessel, causing hydrocarbon vapours to emerge. The hydrocarbon vapour is potentially harmful as it contains various HAPs and VOCs, with high levels of toxicity, which may be inhaled. In the absence of vapour recovery, this hydrocarbon vapour is emitted and pollutes ambient air, deteriorating air quality and potentially affecting human health. Vapour recovery, which can take various forms, attempts to mitigate these emissions and depending on the solution employed, can be very effective and reduce vapour emissions almost entirely (Duarte-Davidson et al., 2001). Different solutions are typically applied to different sectors of the distribution chain. Thus, for example, fuel pumps in the United States have vapour recovery nozzles placed on them to decrease vapour release during refuelling.

However, in South Africa, vapour recovery has only been partially implemented, with complete vapour recovery being implemented only at refinery road tanker loading depots. As yet, there is no vapour recovery in the downstream logistics chain (tanker to service station storage vessel; and fuel pump to vehicle tank), in South Africa. This is of major concern, as fulltime forecourt attendants are employed at service stations across the country, as they are in many developing countries. Thus, their exposure to petrol and diesel during transfer from tanker to forecourt storage vessels and during refuelling of cars, buses and trucks, may be higher than in countries that have vapour recovery systems throughout the logistics chain.

Fuel attendants in the country refuel vehicles with both diesel and petrol. However, studies have mainly focused on petrol vapours and resultant health effects, as diesel has a much lower volatility than petrol. Thus exposure to VOCs from diesel is considered to be considerably lower than that from petrol. However, continuous exposure to VOCs, especially in ‘hot spot’ areas where concentrations of VOCs may be augmented, such as diesel refuelling bays, may still be of concern. Thus, the focus of this thesis will be on diesel service stations only.

## 1.2 Regulation and fuel usage

In South Africa, fuel levies (such as an environmental levy) exist. Thus, consumers pay a premium to counteract harm to the environment. The levies and government taxes are regulated, and thus, fuel companies have restrictions on pricing. There are four main fuel companies in South Africa who supply both petrol and diesel to all retail garages. These are BP, Shell, Engen and SASOL. Fuel types in the country include unleaded petrol, i.e. 93-unleaded and 95-unleaded (93-unleaded petrol is only available in the interior of South Africa and not in coastal areas); LRP, i.e. lead replacement petrol, for older vehicles; and three variants of diesel. These are ultra-low sulphur diesel (10 ppm), which was introduced in 2013; low sulphur diesel (50 ppm); and standard (heavy-end) diesel (500 ppm). Each of the fuel companies enhance their fuel by adding specific additives, which further changes the composition of the fuel, making monitoring and regulation difficult, but imperative.

Over the past ten years, fuel has been regulated more forcefully by governments worldwide. Many governmental organisations have realised the effects of fuel emissions on air quality, human and environmental health and have placed different restrictions on fuel. One such restriction has been by the European Union (EU), where they have regulated the amount of benzene in petrol (Pavlova and Ivanova, 2003). As of 2010, only 1% (v/v) benzene is allowed in petrol. However, despite the increasing awareness around the harmful VOCs released from fuel, the realisation is centred on petrol, and diesel fuel is still under-regulated.

According to the South African Petroleum Industry Association (SAPIA, 2008), the majority of diesel usage in South Africa (59%) is at retail garages (Figure 1.2), where fuel pump attendants are employed. Public transport buses, which are analysed in this study, fall within two sectors of the South African market: that of retail garages (39%) and public transport (2%), as public buses are refuelled at retail garages, and are not captured independently. It should be noted that public buses utilise 500 ppm sulphur diesel, and have fuel tanks almost 3-4 larger that of a regular vehicle. Thus, vapours from refuelling may be more pronounced. In addition to this, emissions from exhaust fumes will also significantly increase levels of VOCs in surrounding areas.

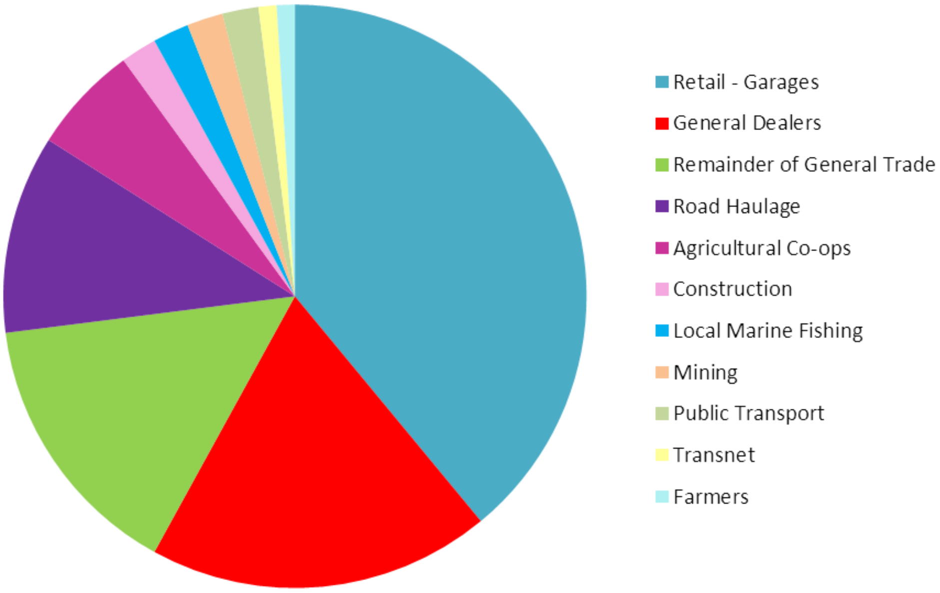

**Figure 1.2: The diesel usage in percentage by different sectors in South Africa as noted in 2008, by the South African Petroleum Industry Association (Source: after SAPIA, 2008).**

Studies have analysed effects from petrol vapour emissions and fumes (Askari et al., 2005; Chang et al., 2009; Das et al., 1991; Gonzalez-Flesca et al., 2002; Hein et al., 1989; Karakitsios et al., 2007a; Keretetse et al., 2008; Kitwattanavong et al., 2013; Majumdar (neé Som) et al., 2008; Morales Terrés et al., 2010; Oesch et al., 1995; Onunkwor et al., 2004; Pandya et al., 1975; Pinedo et al., 2012; Rekhadevi et al., 2010; Rushton et al., 2014; Shin and Kwon, 2000; Singh et al., 2013; Udonwa et al., 2009), while other studies have analysed diesel particulates, and related it to health (Burtscher, 2005; Carvalho-Oliveira, 2005; Chen et al., 2007; Crump, 2014; Ferreira et al., 2008; Nightingale et al., 2000; Stayner et al., 1998; World Health Organization, 2012).

However, no studies, either in South Africa or worldwide, have focused on the release of VOCs from diesel fuel and analysed the potential effects on health, despite full time forecourt attendants being employed. Additionally, as mentioned earlier, diesel fuel is unregulated, and thus levels of benzene in particular are not monitored or controlled, which may pose severe health risk consequences to employees and the public alike. Furthermore, with a significant increase in diesel fuelled vehicles (Figure 1.3), with the wide public impression that diesel is a 'cleaner fuel' (as compared to petrol fuelled cars), there is a significant increase in diesel-fuelled motor vehicles in the country.

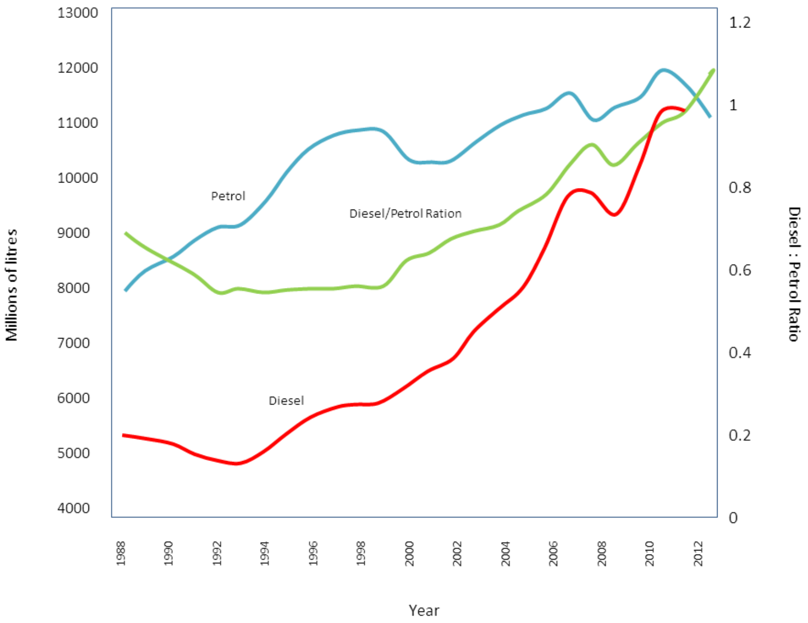

**Figure 1.3: Petrol and diesel consumption in South Africa from 1988 – 2013. South Africa utilised approximately 11.2 billion litres of petrol and 11.9 billion litres of diesel during 2013. There was a marginal decrease in in petrol consumption (2.1%), while diesel displayed a 0.3% increase in consumption, from the previous year (Source: after SAPIA, 2008).**

## 1.3 Air quality and legislative practices in South Africa

Despite diesel fuel not being regulated specifically, air quality, public health and occupational safety are governed by means of a number of legislative Acts in South Africa, including but not limited to:

- Customs and Excise Act (Act 91 of 1964)

  *(Specifically relates to levies and government taxes associated to petrol, diesel and bio-diesel fuels)*
- Hazardous Substances Act (Act 5 of 1973)

  *(Relates to the toxic and/or flammable nature of compounds)*
- Environment Conservation Act (Act 73 of 1989)

  *(Relates to the protection and controlled utilization of the environment, specifically with regards to environmental pollution)*
- Occupational Health and Safety Act (Act 85 of 1993)

  *(Relates to the protection of persons at work against hazards to health and safety arising in connection with work-related activities)*

- Air Quality Act (Act 39 of 2004)

  *(Relates to regulating air quality in order to protect the environment by providing reasonable measures for the prevention of pollution and ecological degradation).*

Previous studies on gas refuelling practices and concentration levels of HAPS around the world have focused on the polluting potential of petrol stations, and oil and petroleum plants polluting air and water; and seasonal and diurnal variations in VOC concentrations in urban air and its effects on human and environmental health (e.g. Caprino and Togna, 1998; Chauhan et al., 2014.; Han and Naeher, 2006; Porta et al., 2009; Smith, 1993). However, no studies in South Africa focus on health risks from diesel refuelling stations, or sites with high diesel emissions, despite pump attendants being employed on a full time basis. Of major concern, at the forecourt from diesel refuelling, is the release of six aromatic hydrocarbons, namely benzene, toluene, ethyl-benzene, o-, m- and p-xylene (BTEX). According to the United States Environmental Protection Agency (US EPA), “BTEX is a term used for benzene, toluene, ethyl-benzene, and xylene- volatile aromatic compounds typically found in petroleum products, such as gasoline and diesel fuel” (US EPA, 2010). Thus, the following sections will focus on the health risks associated with BTEX, and a review of risk assessments associated with BTEX from various countries will be analysed.

## 1.4 Health risks associated with BTEX

BTEX compounds are emitted from various sources, and concentrations are generally higher in urban air-sheds, due to vehicular traffic, industrial activities and chemical manufacturing plants being more active in these areas. Benzene, toluene, ethyl-benzene and xylenes, which are all aromatic hydrocarbons, are found naturally in gasoline, crude oil and diesel fuel, and may be included as additives to gasoline to enhance fuel performance. In a study undertaken by SAPIA (2008), it was noted that although benzene specifically is a matter of concern in petrol, PAHs need to be continually monitored in diesel. Furthermore, it was stated that PAHs are predominantly present in the heavier ends of diesel, such as motor vehicles fuelled with 500 ppm diesel. However, personal exposure to these aromatic hydrocarbons in occupational settings, such as diesel refuelling stations has not been high on the country's agenda, even though exposure to these PAHs has shown to increase one's risk of possible health-related side effects (SAPIA, 2008). Thus, each of the compounds potential health related effects will be discussed briefly.

### 1.4.1 Health risks associated with benzene

Benzene ($C_6H_6$) in its native state is a colourless, flammable liquid, with a relatively sweet smelling odour. However, benzene is classified as a level 1 carcinogen by the World Health Organization (WHO) (World Health Organization, 2012). This means that benzene has been found to be a known human carcinogen, and thus the WHO state that there should be minimal exposure to this compound. According to the ATSDR (2007a) acute exposure to $C_6H_6$ may result in central nervous system (CNS) disorders (including drowsiness, dizziness, headaches, tremors, convulsions), pulmonary disorders (including increased heart rate), gastrointestinal disorders (including vomiting), and may even result in death. Chronic exposure to this compound may impact blood system functions, and lead to leukaemia (cancer of the blood).

### 1.4.2 Health risks associated with toluene

Toluene ($C_7H_8$), in its native state is a colourless, highly flammable liquid (ethyl-benzene and all three isomers of xylene have the same characteristics), and has a similar sweet odour as that of benzene. According to the ATSDR (2000) acute exposure to the compound can cause intoxication, and may cause unconsciousness, leading to death. $C_7H_8$ may also irritate the respiratory tract. Chronic exposure to the compound may result in CNS depression (including mild symptoms such tiredness, confusion, memory loss and impaired colour vision). However, symptoms may cease when exposure is discontinued.

### 1.4.3 Health risks associated with ethyl-benzene

Ethyl-benzene ($C_8H_{10}$) has an odour like that of gasoline. Acute exposure causes CNS depression (eye irritations and dizziness) and affects the respiratory tract. Studies in animals have shown that chronic exposure to the compound causes damage to ears (i.e. hearing loss) and kidney malfunction. Ethyl-benzene is also classified as a possible carcinogen (ATSDR, 2007b).

### 1.4.4 Health risks associated with xylenes

Xylene ($C_8H_{10}$) (also known as dimethyl-benzene), has three isomeric compounds (viz. o- p- and m-xylene), and like the other PAHs, has a sweet smelling odour. Acute exposure to xylenes may result in the same effects as described for benzene, excluding any carcinogenic risk (the US EPA have concluded carcinogenic risk of xylene as inconclusive due to insufficient data). However, chronic exposure may cause damage to both the liver and kidneys (ATSDR, 2007c).

## 1.5 Health risk assessments

The US National Research Council defines human health risk assessments (HRA) as “the evaluation of scientific information on the hazardous properties of environmental agents and on the extent of human exposure to those agents” (Moy, 2005). According to the UK Environment Agency (2000), a three step process is necessary for any (health) risk assessment (Figure 1.4). These three steps are:

1. Hazard identification;
2. Exposure assessment and/or dose-response assessment; and
3. Risk characterization.

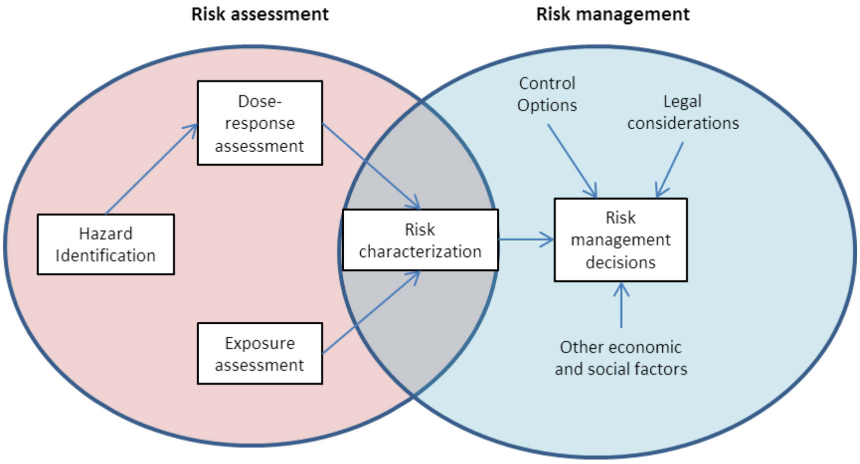

**Figure 1.4: The US National Research Council's Risk Assessment Model, as developed by the United States Environmental Protection Agency (US EPA) (Source: after US EPA, 2010).**

In order to provide the context for this study, and the methodological approach employed, the following definitions related to HRAs are offered:

- Risk- the potential adverse effect that would be caused by a hazard (Colman Lerner et al., 2012).
- Hazard- any chemical, physical or biological agent (or a combination of these), that have the potential to cause harm.

Thus, the nature of the hazard, the potential exposure, occurrence and magnitude of the exposure, and the exposed population characteristics, determine the overall risk. In this manner, a risk assessment is a process undertaken in order to recognise any likely negative effects of exposure. It is a commonly used tool to link environmental exposure to potential human health effects. Due to the characteristics of health risk assessments (being qualitative or quantitative), it has the benefit over other methodologies (such as observational studies or analytical epidemiological studies), in that they are predictive in nature. It uses readily available information, such as exposure data, to quantify health effects of exposure to a certain substance/s. Risk assessments thus, may be conducted over a much shorter time period than other methodologies, making them economically advantageous, yet provides valuable predictions (Askari et al., 2005; Colman Lerner et al., 2012; Kitwattanavong et al., 2013; Moy, 2005; Pinedo et al., 2012).

In recent times, health risk assessments, specifically regarding vehicle exhaust fumes and refuelling stations, have become common. These epidemiological studies include those that have analysed the effects of VOC emission, specifically with regard to BTEX emissions due to their high levels of toxicity. The studies have evaluated the incidence of cancers, birth defects, reproductive disorders, and respiratory diseases (Rushton et al., 2014; Wiwanitkit, 2008).

Other reports have conducted HRAs near areas with high levels of VOCs. These include petrol stations, bus stops, and areas with high traffic movement (such as road tunnels) across Europe and Asia (Alhaji, 2011; Frey et al., 2007; Gonzalez-Flesca et al., 2002; Moore and Figliozzi, 2011; Moore et al., 2012; Murena, 2007). Health risks to passengers using public transport buses were also analysed and, generally, levels of benzene exposure were found to be slightly higher in passengers exposed to diesel particulates (Armas et al., 2012; Chen et al., 2011; Liu et al., 2011; Yu and Li, 2014).

Risk to auto mechanics and painters were also analysed in other studies, as these occupational settings also placed these employees at increased risk to potential health complications from inhalation exposure (Badjagbo et al., 2010; Onunkwor et al., 2004; Udonwa et al., 2009). These studies revealed that auto-mechanics experienced increased risk exposure, as compared to the general population not exposed to VOCs, related to vehicular emissions and particulates. However, the findings revealed that the risk was not as great, as occupational settings such as service station attendants, who experienced greater risks (Colman Lerner et al., 2012).

In addition to this, Porta et al. (2009), concluded that epidemiological studies did not include all limiting factors and confounding issues, such as previous medical illnesses; duration of exposure; level of socio-economic status; sex; year of birth (in children); working conditions; and/or personal habits (such as smoking and drinking). In addition to these limiting factors, single and multi-site analyses provide different results, and sometimes inconsistent findings.

In conclusion, health risk assessments indicate a possible and/or minor risk of cancer, respiratory-related diseases and birth defects of individuals exposed to high levels of VOCs and BTEX, due to environmental and/or occupational surroundings. Studies also indicate that employees of petrol refuelling stations are at risk in terms of respiratory-, dermatological-, and neurological-related diseases, as well as increased chance of developing carcinomas (Smith, 1993). Thus, in order to not only manage emissions at refuelling stations, but to protect individuals, health risk assessments are vital. Additionally, a 'mixed-methods' approach is necessary in order to exclude bias and reduce limiting factors within the study.

## 1.6 Aims and objectives of the study

No health risk assessment on diesel refuelling stations has been conducted in South Africa, or in any developing country, to date. Thus, this research aimed to evaluate whether emissions at a bus depot in Johannesburg, South Africa, has the potential to cause adverse health effects to employees at the site, by conducting an HRA.

The main objectives of this research were:

1. To determine the concentrations of BTEX at a bus diesel refuelling bay,
2. To ascertain if BTEX emissions have a diurnal variation,
3. To determine whether BTEX emissions exceed national and/or international guidelines and occupational limits, and the implications thereof, and
4. To investigate if BTEX concentrations have the potential to affect the occupational health and safety of workers at the site.

## 1.7 Study area and site description

The experiment was conducted at a refuelling bay located in Johannesburg, South Africa (Figure 1.5). Johannesburg, located on the interior plateau of the country, experiences cold, dry winters, with winter temperatures (June, July and August) ranging from −3 to 19 °C.

The bus depot belongs to a government owned entity, and services the public transport sector, throughout the Johannesburg metropolis. The bus depot is open daily, however, the refuelling bay and adjacent workshops, offices and repair centres are only open during weekdays (Monday – Friday), 07:30-15:30.

The refueling bay is under cover, and has four doors, 3 m high on either end of the 30 m bay. There are four refueling pumps, centrally located, where two buses can be refueled simultaneously. The depot itself can accommodate 400 parked buses, and refuels on average 85 buses daily. This equates to approximately 6,700 liters of 500 ppm Sulphur diesel being pumped per day during the work hours. There are four full time employees at the refuelling bay, and over 50 employees within the bus depot in adjacent buildings. In close proximity to the refueling bay (Figure 1.6a) is a large enclosed workshop where maintenance and repairs take place (Figure 1.6b).

At the study site, pump attendants wear protective gloves when refueling the buses; however, these are removed when busy with other activities while refuelling (Appendix i). Additionally, in South Africa no rubber hood is used over the delivery pump, as is the practice in the USA to reduce emissions (Udonwa et al., 2009). There is very little ventilation, despite bay doors allowing a small amount of airflow to be present, as all four extractor fans located near the ceiling of the bay were non-operational throughout the monitoring period, thus exhaust fumes are not removed rapidly (bus engines idle during refuelling to save costs), and may significantly contribute to recorded emissions. Furthermore, 36% of buses at the depot failed the smoke tests conducted by the company. The smoke test indicates whether exhaust fumes released by the bus are within the acceptable range (as determined by the bus manufacturers), as well as if burning of the fuel is occurring correctly. This implies that exhaust fumes within the refueling bay may be further intensified. Spills are also not well managed on site, which further deteriorates the air quality (Appendix ii).

This situation however, is not unique to this site. Government-owned public bus companies, throughout South Africa, experience similar conditions. There are 12 urban bus diesel-refuelling stations across the country, all under-cover, and have little to no ventilation and/or filtration systems in place. Additionally, six of these refuelling bays service outdated buses that do not pass the smoke tests, but are still in operation, due to lack of funds. Moreover, all the government-owned bus depots in the country have full time employed fuel pump attendants. Thus, results and findings from this study can be generalized, and recommendations are applicable in all bus diesel-refuelling stations, and other developing countries, especially in Africa. However, when contacted to conduct monitoring research at these sites, permission was denied.

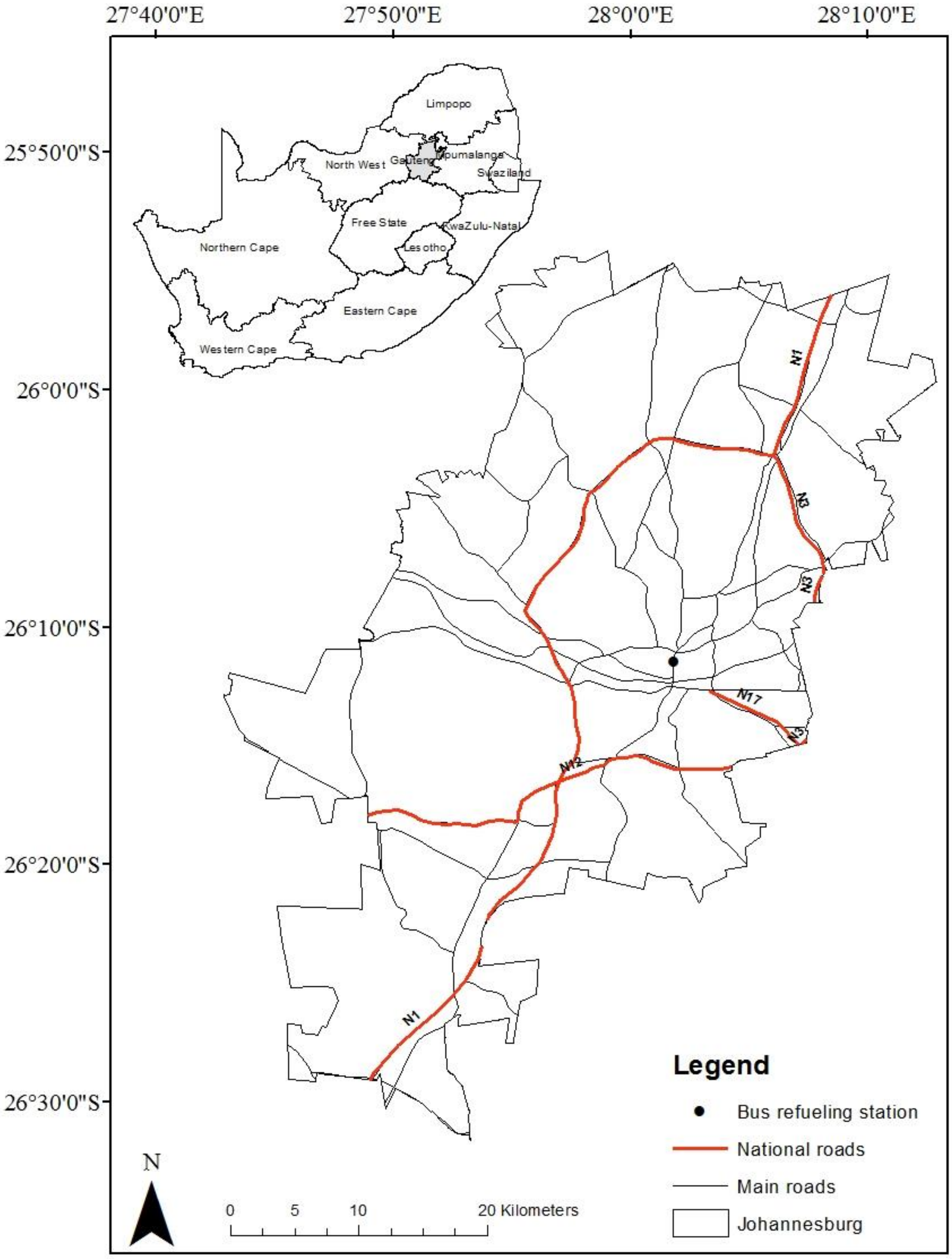

**Figure 1.5: The location of Johannesburg situated within the province of Gauteng, South Africa (top left), and the refuelling bay located within the CBD of Johannesburg (GPS co-ordinates: 26.1908° S, 28.0303° E) (map data provided by South African National Space Agency).**

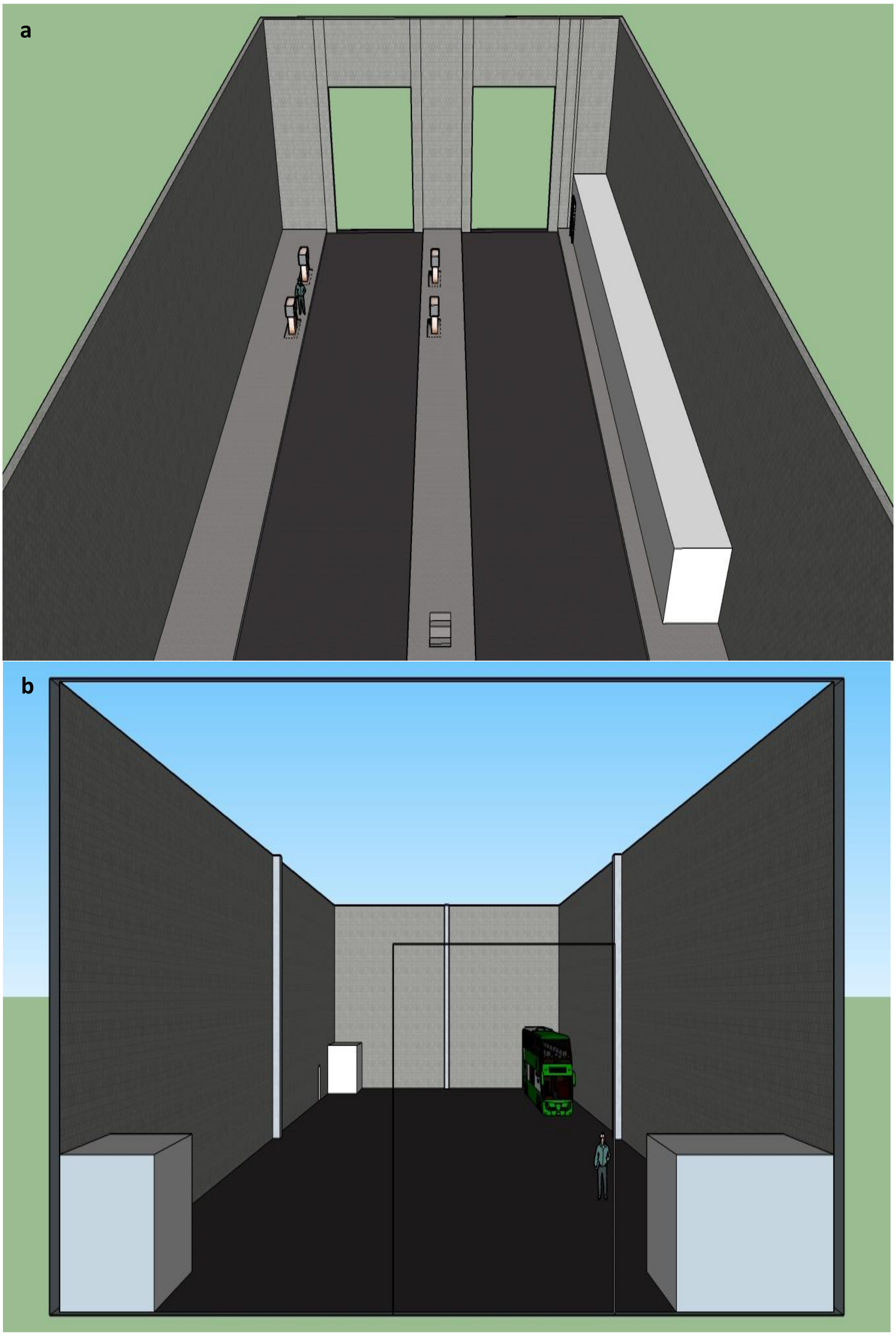

Figure 1.6: Sketches displaying the (a) fuel bay, and (b) workshop on site. The fuel bay is adjacent to the workshop at the bus depot (Overhead roofs and front walls have been made transparent for viewing purposes. Offices displayed as white boxes).

# 1.8 Methodological approach

## 1.8.1 Continuous sampling strategy

Data collection was carried out at an urban diesel-refuelling bay under normal operation conditions throughout the week for a three-month period from June-August 2013 inclusively (i.e. winter season). The study period was chosen within the colder, dry season in Johannesburg, when air pollution is commonly higher in the city, and BTEX concentrations are known to be at a maximum (Gallego et al., 2008; Hoque et al., 2008; Schneider et al., 2001; Zalel et al., 2008), which may lead to biases in the data set, i.e. over-estimation compared with annual mean exposure levels. In addition, the short period of monitoring may increase risk of errors. However, a study conducted by Truc and Oanh (2007) followed a similar methodology, with the aim of determining exposure levels. Continuous monitoring at the refuelling bay was carried out for 24 hours, 7 days a week, at 15 minute intervals, during both work and non-work hours (13,300 data points in total); to achieve a comprehensive analysis of daily BTEX concentrations, irrespective of activity or inactivity within the fuel bay.

The original study design intended to carry out a comparative summer campaign, but high summer temperatures in the depot caused major instrumental problems. However, data were successfully collected over a 72 hour period in summer, providing some comparative results for another season (see Chapter 3).

As outlined by Murena (2007), sites that are deemed to have high concentrations of air pollutants can have major implications on human health and, thus, the use of automated systems can be advantageous as continuous data can be collected. For this reason, ambient air was sampled using the Syntech Spectras gas chromatography 955, with a photo ionization detector (PID) (GC955, series 600, using a capillary column AT1 with a 100% dimethylpolysiloxane column packing). Additionally, a PID is particularly useful as it is suited for continuous monitoring and specifically sensitive for aromatic hydrocarbons, even in very low concentrations. The sampling flow was set to sample at 15 minute intervals and sampling height of ambient BTEX was 1.5 m at the center of the fuel bay, with 8 mm tubing and Swagelok fittings used for connecting tubing. Following the methodology of Gallego et al. (2008) and Shin and Kwon (2000), helium was used as the carrier gas in the GC955, set at a pressure of 350 kPa. The instrument has been tested for accordance to the EMC directive 89/336/EMC, test specification EN 50081-1:1991 and EN 50082-2: 1994. The manufacturer stated error of instrumentation is <2%. A gas mixture (1 ppm of each BTEX) in a gas cylinder (provided by AirLiquide) was adopted to identify elution time and chromatographic peaks. Six aromatic hydrocarbons were continuously monitored (viz. benzene, toluene, ethyl-benzene, and xylenes). The monitoring instruments were calibrated before use (calibration was done in the range of 0 to 10 ppb). Quality control checks were conducted during and after the monitoring campaign and a correction factor of 2 ppb and 4 ppb for benzene and toluene were determined, respectively, to counter systematic under-sampling of the instrument. Additionally, 97 data points (<1% of data set) were discarded, as they were distinct outliers.

Additionally, a Davis weather instrument (with data logger CR10X), and a Luft Weather Sensor, with built-in temperature, humidity, wind speed and wind direction sensors, was deployed within the refueling bay, as ambient climatic conditions are known to affect pollution levels (Fenger, 1999). These were mounted at a height of 2 m above ground level inside the station. Temperature, pressure and humidity were recorded every 30 minutes, throughout the entire monitoring period. Temperature recordings were corrected to a ±3˚C, as stated by the manufacturers. Fuel logs were obtained from MetroBus Pty. Ltd (which is the government owned entity that runs the bus service). Fuel logs are maintained at the site, based on fuel filled per bus (in litres), per day.

### 1.8.2 Passive sampling strategy

Sampling and monitoring of passive samplers occurred during the winter (mid-2014), as many studies using both active and passive sampling strategies have established that BTEX concentrations are elevated in winter as compared to other seasons (Gallego et al., 2008; Hoque et al., 2008; Zabiegala et al., 2010; Zalel et al., 2008).

Radiello passive air samplers were arranged in both the refueling bay and workshop, where emissions were considered to be at their maximum. These samplers are considered reliable in both indoor and outdoor environments and, following the European standard (EN 13528-2), the Radiello passive samplers were used to analyze the risk exposure of BTEX in this situation (Pennequincardinal et al., 2005a, 2005b).

Concentrations as low as 2 µg·m$^{-3}$ may be measured with the samplers, with an error not exceeding 0.1 µg·m$^{-3}$ (according to the manufacturers). The samplers were deployed for 14 days as prescribed by the manufacturer. The BTEX passive samplers consist of an absorbing cartridge, which is placed in a micro-porous polyethylene membrane surface (50 mm long micro-porous cylinder; 16 mm external diameter; 300 mg of 40–60 mesh Carborograph 4) (Król et al., 2012; Pennequincardinal et al., 2005a).

Each cartridge was secured to a triangular polycarbonate supporting plate. The sampling plates and cartridges were not placed within protective chambers, as wind speeds were low enough to avoid error (Shoeib and Harner, 2002) (average wind speeds were 2 m.s$^{-1}$ in the refueling bay and workshop, as they are undercover sites). As outlined by Gallego et al. (2008), each sample was labelled and initial and final sampling times were recorded. As soon as the sampling process was over, tubes were returned to their protective containers and sent to a laboratory for analysis (ChemTech Labs, Johannesburg, South Africa). Tubes containing the samples were stored in a dark, cool box. As advised by the laboratory, leaded pencils were avoided as to preclude any contamination of the samples.

### 1.8.3 Risk characterization

As has been shown in many studies, inhalation risk analysis is vital in order to determine the potential exposure of employees (Durmusoglu et al., 2010; Kitwattanavong et al., 2013; Majumdar et al., 2008; Tunsaringkarn et al., 2012). Both cancer risk and hazard risk calculations (associated with the inhalation of air pollutants) were done for employees to evaluate the potential effect of BTEX on human health. Individual calculated cancer risk and hazard risk values were compared with the United States Environmental Protection Agency (US EPA) acceptable standards.

To calculate cancer risk (CR) Equation (1) was applied, while Equation (2) was used to evaluate the non-carcinogenic hazard quotient (HQ):

$$\text{Cancer Risk (CR)} = \text{Lifetime Average Daily Dose (LADD)} \times \text{Slope Factor} \qquad (1)$$

$$\text{Hazard Quotient (HQ)} = \text{Lifetime Average Daily Dose (LADD)}/\text{reference dose} \qquad (2)$$

Inhalation slope factor (SF) [benzene 0,0273 $(\mathrm{mg/kg/day})^{-1}$] and reference dose (RfD) standard values were used [benzene 0,00855 $\mathrm{mg{\cdot}kg^{-1}{\cdot}day^{-1}}$, toluene 1.43 $\mathrm{mg{\cdot}kg^{-1}{\cdot}day^{-1}}$ and xylenes 0,029 $\mathrm{mg{\cdot}kg^{-1}.day^{-1}}$] (Durmusoglu et al., 2010; Edokpolo et al., 2014).

To calculate the Lifetime Average Daily Dose (LADD) of employees, Equation (3) was utilised:

$$\mathrm{LADD = (C.CF.IR.EF.ED)/(BW.AT)} \tag{3}$$

where C is the contaminant concentration (average concentrations used from passive samplers) ($\mu g/m^3$); CF is the conversion factor (1 mg/1000 µg); IR is the inhalation rate (US EPA standard) (20 $m^3$/day); EF is the exposure frequency (days/year); ED is the exposure duration (years); BW is body weight (US EPA standard) (70 kg); and AT is the averaging time (exposure averaged over life time/average life expectancy for male and female) (days) (for specific values refer to Appendix iii).

The depot operations manager provided demographic data, in order to provide information pertaining to individual input variables such as age, sex, exposure frequency and exposure duration. Where data was limited, US EPA standard values were used for body weight (70 kg for males and 60 kg for females) and inhalation rate (20 $m^3$/day) (Durmusoglu et al., 2010). In order to calculate the exposure frequency, standard values were used (all employees worked a standard five days, eight hours per day and received a minimum of 21 days leave per annum). More detailed evaluation of confounding factors, such as smoking habits and home conditions, was beyond the scope of this study.

### 1.8.4 Study sample

The occupationally exposed group consisted of fuel bay attendants (FBA) from the refueling bay (n = 4) and diesel auto mechanics (AM) from the bus workshop (n = 16). Only full time employees were considered for this study. The demographic information provided by the bus company is illustrated in Table 1.1.

**Table 1.1: Demographic data of participants in the fuel bay attendants (FBA) and auto-mechanics (AM). Data supplied by bus operating company.**

| Participant ID | Workplace | Gender | Smoker | Age | Employment Duration |
|---|---|---|---|---|---|
| | | | | (years) | (years) |
| FBA1 | Fuel Bay | Male | No | 27 | 5 |
| FBA2 | Fuel Bay | Male | No | 45 | 10 |
| FBA3 | Fuel Bay | Male | No | 59 | 33 |
| FBA4 | Fuel Bay | Male | Yes | 56 | 37 |
| AM1 | Workshop | Male | Yes | 27 | 1 |
| AM2 | Workshop | Male | Yes | 35 | 1 |
| AM3 | Workshop | Male | No | 25 | 2 |
| AM4 | Workshop | Male | No | 26 | 2 |
| AM5 | Workshop | Male | No | 24 | 2 |
| AM6 | Workshop | Male | No | 25 | 3 |
| AM7 | Workshop | Male | No | 29 | 4 |
| AM8 | Workshop | Male | No | 36 | 5 |
| AM9 | Workshop * | Female | No | 47 | 8 |
| AM10 | Workshop * | Male | Yes | 40 | 10 |
| AM11 | Workshop | Male | No | 41 | 10 |
| AM12 | Workshop | Male | Yes | 51 | 11 |
| AM13 | Workshop | Male | No | 40 | 16 |
| AM14 | Workshop | Male | No | 38 | 16 |
| AM15 | Workshop | Male | No | 49 | 28 |
| AM16 | Workshop * | Male | No | 63 | 41 |

* Employee based within an enclosed office inside the workshop.

The majority of the employees at the site were male (only one auto-mechanic at the workshop was female). Overall, the majority of the employees did not smoke (75%). As can be seen in Table 1.1 there is a very wide range of employee ages (24 to 63 years) and exposure duration (from 1 to 41 years). All employees worked a standard 8 hours per day, 5 days a week.

### 1.8.5 Limitations to methodological design

The methodological design had subsequent flaws that should be noted. A shortcoming of subsequent chapters refers to the analyses of the data regarding the continuous sampling strategy using the GC955. The manuscripts only refer to two isomers of xylenes. This however is incorrect as m- and p-xylene co-elute in the GC955. Thus, where reference is made to only p-xylene, it should state both (unfortunately this error could not be re-worded as the papers have been published).

In addition to this, due to instrumentation failure, there were slight gaps in data. However, with the large data set acquired, statistical validation was still possible. The GC955 was also problematic, and could not function in high temperatures, and thus the study was centred on winter monitoring campaigns. However, previous research iterates that BTEX concentrations are highest in winter (Gallego et al., 2008; Hoque et al., 2008; Król et al., 2012; Schneider et al., 2001; Zalel et al., 2008), hence results are still important for analyses. (Despite this, a relatively short summer campaign has been included to further re-iterate this point in Chapter 3.) Another restriction was that the site was undercover, and thus variations in climatic conditions were not as variable as outdoor environments. This however is not a point of concern when looking specifically at these types of refuelling stations in the country, as most bus refuelling stations operate under very similar conditions.

Another constraint was with regard to the use of only Radiello samplers, in an area with high concentrations of BTEX. It has been mentioned that Radiello samplers should be used with caution in work environments with high concentrations of VOCs (as this may lead to under-sampling) (Pennequincardinal et al., 2005a, 2005b), however, the use of personal monitors was not possible for two reasons. The first is that the Radiello samplers were more economically viable, and second, permission to use personal monitors at the site was not granted. The bus company wanted to avoid causing any possible anxiety amongst their employees. Thus, passive samplers were employed as the alternative, despite its limitations. Likewise, it should be noted that due to the concentrations being so high, there could have been incidences of back diffusion occurring in which higher concentrations may be erroneously recorded. In retrospect, the Radiello passive samplers should have only been deployed for 7 days (and not 14 days), and multiple sampling campaigns should have been conducted, however, due to financial constraints, this was not possible.

Overall, the combined approach of these methodologies (i.e. continuous sampling, passive sampling, and risk characterisation using mathematical calculations), despite their respective limitations, have postulated, if nothing else, proxy estimates of potential health risk of employees, which can facilitate future research.

## 1.9 Structure of the thesis

This thesis consists of the present introductory chapter (Chapter 1), a general discussion and conclusion chapter (Chapter 5) and the main body comprising of experimental chapters (Chapters 2-4) (see Table 1.2). The experimental chapters are publications (accepted or under review). All three chapters aim to quantify BTEX concentrations at the site (objective 1); with Chapter 2 focusing on diurnal variation of BTEX concentrations; Chapter 3 aiming to determine whether BTEX emissions exceed national and/or international occupational limits, and the implications thereof; while Chapter 4 investigated the potential occupational health and safety of workers at the site. Chapter 5 is a general discussion and providing overarching conclusions, drawing together the main findings. Each chapter has been formatted to the specific journal/publishers' requirements, thus each of these chapters include substantial background information, a detailed methodological approach, as well as an abstract and a reference list. The tables and figures have been numbered for each journal paper and the pages of the thesis are numbered in chapter sequence. References are provided in each chapter.

Table 1.2: Overview of experimental chapters, presented as manuscripts.

| Chapter | Title | Aim | Objectives | Instruments | Data details |
|---|---|---|---|---|---|
| Two | BTEX concentrations influenced by external factors at a diesel-refuelling station in Johannesburg, South Africa | To analyse the two main factors that are influential on fluctuations of ambient BTEX concentrations | To determine the total volume of fuel dispensed and ambient temperature in relation to BTEX concentrations. | Spectras Gas Chromatography 955 VOC analyser; Davis meteorological weather station; fuel logs. | 3 months winter data (19 June- 30 August 2013).<br>Daily and 15 minute BTEX concentrations analysed; daily temperature data analysed. |
| Three | Assessment of occupational exposure to BTEX compounds at a bus diesel-refuelling bay: A case study in Johannesburg, South Africa | To evaluate emission characteristics at a diesel refuelling station | To collect air concentration data from diesel refuelling and exhaust emissions of BTEX; to evaluate whether these chemical concentrations are above legal standards, both national and international; and to investigate the variation of BTEX concentrations during a typical day on site. | Spectras Gas Chromatography 955 VOC analyser | 3 months winter data (19 June- 30 August 2013); 72 hours summer data (01-03 January 2014).<br>Analysis made on 8 hour time weighted averages and BTEX species |
| Four | Occupational exposure of diesel station workers to BTEX compounds at a bus depot | To conduct a health risk assessment of BTEX at a diesel service station for public buses | To perform site-specific health risk analysis of occupational exposure to BTEX | Radiello passive samplers; Luft Weather Sensor | 14 days in winter (02-15 July 2014).<br>Demographic data of employees provided by operations manager.<br>Analysis using mathematical calculations, in relation to BTEX concentrations. |

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# CHAPTER TWO: BTEX CONCENTRATIONS INFLUENCED BY EXTERNAL FACTORS AT A DIESEL-REFUELLING STATION IN JOHANNESBURG, SOUTH AFRICA

**R. Moolla**, C. J. Curtis, and J. Knight 2014: BTEX Concentrations Influenced By External Factors At A Diesel-refuelling Station in Johannesburg, South Africa, *The Sustainable City IX, Volume 2*: 1459-1467 (DOI: 10.2495/SC141232).

**Abstract**

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# BTEX concentrations influenced by external factors at a diesel-refuelling station in Johannesburg, South Africa

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## Abstract

Public transport systems in Johannesburg, South Africa, rely on a large number of diesel-powered buses. These buses are fuel economical and durable. However, filling station attendants, bus drivers and the public are exposed to the diesel fuel and fumes associated with them. Fuel attendants are exposed to diesel exhaust fumes, as well as emissions from fuel pumps on a daily basis, and are at risk to adverse health effects associated with inhalation of volatile organic compounds (VOCs) released. The VOCs released include benzene, toluene, ethyl-benzene and xylenes (BTEX), which have a high level of toxicity. Studies relating to the concentrations of BTEX at diesel stations are limited, as most studies focus on petrol refuelling stations. Thus, analyses of these concentrations are significant within developing countries whose transport systems rely on diesel-powered buses, and where public health measures are often less rigorously enforced. As this research falls within a larger study relating to the health impact of BTEX on fuel attendants at a diesel-refuelling bay, an initial study was undertaken to analyse the two main external factors that are influential on fluctuations of ambient concentrations. Thus, an analysis of total volume dispensed, and ambient temperature at the station, both affecting the concentrations of BTEX released, was conducted. It was established that $BTEX_{total}$ concentrations were positively correlated to the volume of diesel dispensed daily and inversely correlated to temperature. Additionally, ethylbenzene and o-xylene indicated a positive correlation with volume of fuel dispensed, while toluene and p-xylene were negatively correlated to temperature.

*Keywords: benzene, toluene, ethylbenzene, xylenes, diesel, temperature.*

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# 1 Introduction

Diesel exhaust fumes, released from motor vehicles, buses, locomotives and other motorized machinery, has three major groups of sources (i.e. mobile sources, stationary sources and stationary point sources) [1, 2]. In addition, vapours are released from diesel fuel at refuelling bays and filling-garages. These vapours include various volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and particulates [3–5]. However, many studies have focused on specific VOCs, namely the BTEX group (benzene, toluene, ethyl-benzene and xylenes) which are released by petrol and diesel fuels. The amount of BTEX concentrations released from the fuel can vary according to the composition and additives in the fuel, as some additives may increase benzene concentrations. In addition, the ways in which the diesel fuel is used at specific sites, such as parking or refuelling bays, can affect the concentrations, as well as ambient climatic conditions.

Epidemiological studies have shown that inhalation of fuel vapours can be hazardous to human health, specifically to fuel attendants [6–12]. BTEX in general have been shown to be associated with a range of health complications such as cardiopulmonary disease, lung, liver, and kidney diseases [6, 7, 10]. The inhalation of benzene has also been linked to adverse teratogenic effects [3, 13, 14]. In recent years diesel exhaust has been suggested as a probable human carcinogen [1, 12, 15, 16], however, very little research specifically on diesel vapour inhalation has been conducted.

BTEX concentrations and associated inhalation thereof vary due to several factors such as total fuel-dispensed, number of shifts and/or hours per shift of employees. Hein *et al.* [6] stated that exposure to fuel vapours can be markedly influenced not only by total volume of fuel dispensed by the attendants during each shift, or length of each shift, but also by changes in atmospheric temperature, ventilation and/or concentration of benzene in the fuel. However, very little literature is available on the effects of temperature and pressure, and/or total volume of fuel dispensed linked to fluctuations in concentrations of BTEX, specifically in diesel fuel. Thus, the main aim of this study was to investigate whether daily BTEX concentrations in diesel varied according to fuel dispensed in one refuelling bay, at a metropolitan bus company in Johannesburg, South Africa. In addition, the study evaluated whether atmospheric temperature also played a role in fluctuations and peaks of concentrations, at the indoor diesel-refuelling bay.

# 2 Background

In South Africa, public transport systems rely on a large number of diesel-powered buses as they are fuel economical and durable. However, filling station attendants, bus drivers and the public are exposed to the diesel fuel and fumes associated with them. Fuel attendants are exposed on a daily basis to not only diesel exhaust fumes, but also emissions from fuel pumps. This exposure places them at particular risk to adverse health effects associated with inhalation of these BTEX, which have a high level of toxicity. There have been significant mechanisms introduced in South Africa to reduce emissions in the fuels, which include a move away from leaded petrol and high content sulphur diesel fuels. LRP (lead replacement petrol), 95-unleaded petrol, 97-unleaded petrol, and 10- and 50-ppm diesel are now the alternatives in South Africa, which are meant to have lower rates of harmful emissions. However, despite these advances, diesel fuel still poses significant risk to fuel attendants.

### 2.1 Site description

In this study, an indoor diesel-refuelling bay was monitored. The fuel bay is located in the hub of Johannesburg and supplies 50ppm fuel to 400 diesel-powered buses. The refuelling bay has four pumps, manned by two full time employees (with additional employees conducting various other duties in and near the bays). The refuelling bay is 30m long, with 3.5m access doors on either end of the bay. Buses are refuelled during working hours (07:00–15:30), Monday to Friday.

## 3 Materials and method

### 3.1 Sampling strategy

Continuous in situ measurements of benzene, toluene, ethyl-benzene and o- and p-xylenes were obtained using the SYNSPEC Spectras Gas Chromatography 955 VOC analyser. Ambient air was sampled at a 1.5m height, at the diesel filling pumps, measured continuously at 15 minute intervals, for the entire winter period (June, July and August). The analyser was calibrated prior to the testing period (calibration was done in the range of 0 to 18$\mu g/m^3$), and a correction factor of 2ppb and 4ppb for benzene and toluene were used, respectively [17]. Helium gas was used as a carrier gas in the GC955 analyser as it is an inert gas and thus safe at the diesel refuelling bays. The winter season provided the conditions for the GC 955 analyser to operate at its optimal, as the pilot study revealed the instrument was non-functional in hot summer temperatures, due to levels of BTEX being too high and causing errors and malfunction to the software. Additionally, the winter season experiences a prevailing high pressure system, which allows the gaseous vapours to accumulate over the site.

Additionally, a Davis weather instrument (with data logger CR10X), was mounted at a height of 2m above ground level inside the station. Temperature, pressure and humidity were recorded every 30 minutes, throughout the entire monitoring period. Temperature recordings were corrected to a ±3°C error. Fuel logs were obtained from MetroBus Pty. Ltd (which is the government owned entity that runs the bus service). Fuel logs are maintained at the site, based on fuel filled per bus (in litres), per day.

### 3.2 Statistical analysis

An interaction term between concentration and fuel dispensed/weather parameters/work and non-work days was included in the analysis. Daily and quarter-hourly averages of BTEX concentrations were used in order to be comparable to daily fuel dispensed and temperature records, respectively. Following the methodological approach of Keretetse *et al.* [9] the level of significance was set at 5%. Statistical analysis was undertaken with the aid of the SPSS 20.1 statistical software. Variables were tested for normality, and non-normally distributed data were analysed using non-parametric tests (viz. Wilcoxon sign rank and Spearman's correlation tests).

## 4 Results and discussion

### 4.1 The influence of fuel dispensed on daily BTEX concentrations

Figure 1 presents concentrations of the $BTEX_{total}$ concentrations obtained at the site for the duration the monitoring period, showing a statistically significant decrease of concentrations on non-workdays (F=1.953, p=0.0403). This was also apparent in a study conducted by Keretetse *et al.* [9], where a significant relationship was also found between the levels of benzene, toluene and total VOCs, and the volume of the petrol sold. However, it is notable that this was not the case in the diesel-refuelling bay in this study.

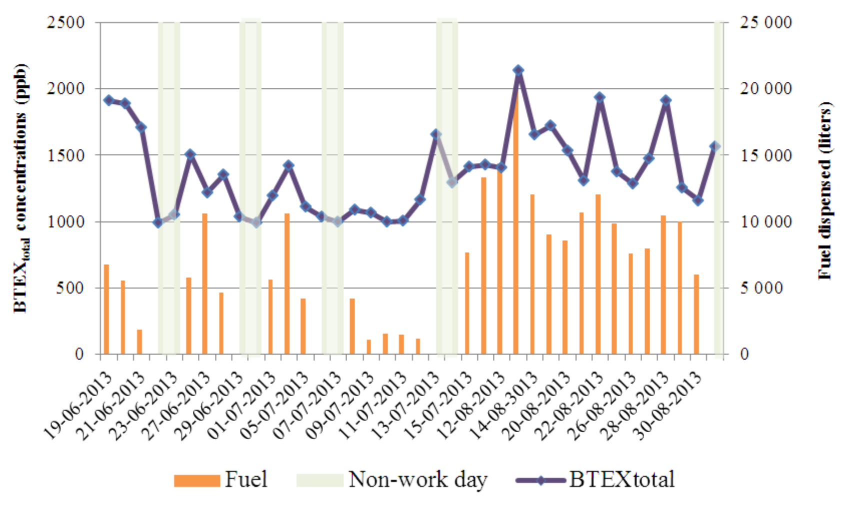

Figure 1: Daily average of ambient $BTEX_{total}$ concentrations as compared to the total volume of diesel dispensed daily at the site during the monitoring period. The shaded area indicates non-work days when the refuelling bay is closed.

Despite the trend for $BTEX_{total}$, benzene and toluene concentrations (Figure 2a, b) are not dependent on fuel consumption. This could be attributed to the fact that, according to Heeb *et al.* [18], both of these aromatic hydrocarbons can undergo prolonged photochemical decay, and this may result in increased rates of ambient benzene and toluene in diesel fuels. In addition, Rasmussen and Khalil [19] found that atmospheric benzene levels were highest during winter, which may explain the high levels recognised at the station, as the study period was during this season.

Ethylbenzene and p-Xylene on the other hand (Figure 2c, d) are positively correlated to the volume of diesel dispensed daily (p=0.478 and 0.547, respectively (Table 1)). This indicates that as volume of fuel-dispensed increases, concentrations also increase. This finding is also in line with those of other studies (e.g. Keretsetse *et al.* [9]), indicating that volume of fuel sold significantly influences ethylbenzene and xylene concentrations.

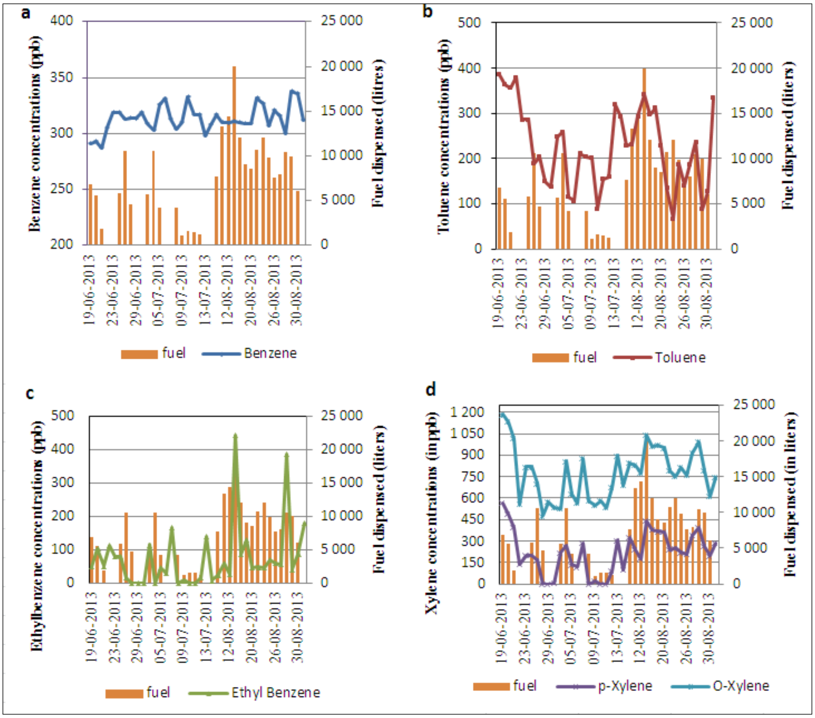

Figure 2: Ambient benzene (a), toluene (b), ethylbenzene (c) and xylene (d) concentrations as compared to fuel dispensed daily at the station, for winter (JJA) 2013.

## 4.2 The influence of temperature on hourly BTEX concentrations

A significant result in this study was that $BTEX_{total}$ concentrations were negatively correlated (p=-0.555) to temperature for the entire monitoring period (Table 1). This can be clearly seen in Figure 3, where daily ambient $BTEX_{total}$ concentrations and ambient temperature variations are compared. However, if the findings are

Table 1: Correlation between BTEX concentrations and the influential factors considered. (r indicates the linear correlation coefficient between two variables, and p indicates the level of significance).

| **VOC** | **n** | **Volume of diesel sold** | | **Temperature** | |
|---|---|---|---|---|---|
| | | **r** | ***p*** | **r** | ***p*** |
| **Benzene** | 96 | 0.113 | 0.113 | 0.197 | 0.197 |
| **Toluene** | 96 | 0.045 | 0.045 | 0.013 | -0.816[b] |
| **Ethylbenzene** | 96 | 0.478 | 0.478[a] | 0.250 | -0.221 |
| **o-Xylene** | 96 | 0.139 | 0.139 | 0.390 | -0.670[b] |
| **p-Xylene** | 96 | 0.547 | 0.547[a] | 0.656 | -0.007 |
| **$\text{BTEX}_{\text{total}}$** | 96 | 0.553 | 0.553 [a] | 0.456 | -0.555[b] |

[a] Positive significance
[b] Negative significance

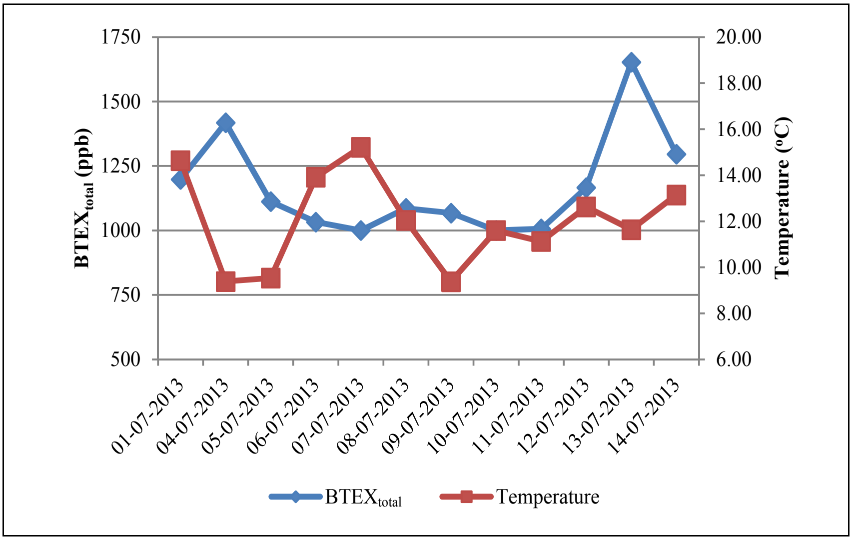

Figure 3: Daily ambient $\text{BTEX}_{\text{total}}$ concentrations as compared to ambient temperature at the station during a two-week period in the winter season.

evaluated against the study conducted by Keretsetse *et al.* [9], temperature was positively correlated to BTEX concentrations. One reason for this could be that diesel and petrol emissions may react differently to fluctuations in temperature, and thus, concentrations of diesel are negatively correlated to temperature.

This is further illustrated when hourly concentrations are contrasted to hourly temperature changes, on a typical winter's day (Figure 4). However, the fluctuations noted may be attributed to a time lag offset in the data, as well as indicative that other factors may be involved (such as humidity, wind and/or atmospheric pressure).

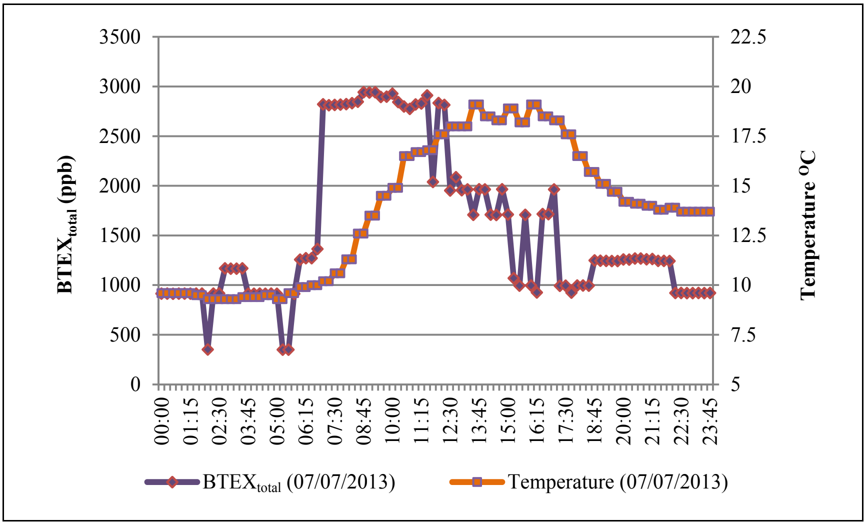

Figure 4: Hourly ambient BTEX$_{total}$ and ambient temperature variations on a single average winter's day during July.

Toluene and o-xylene observe the same behavioural pattern of a negative correlation to temperature (Table 1). A noteworthy finding was that benzene concentrations were not affected by fuel dispensed (Figure 2a) or changes in temperature. However, it was found that peaks in concentrations were closely related to the timing of sunrise daily (06:30–06:45), and this inevitably influenced the BTEX$_{total}$ concentrations to peak at sunrise (Figure 4).

Further investigation into this phenomenon is imperative, as studies have shown that relative humidity, atmospheric pressure and/or wind speed, besides temperature, may contribute to fluctuations in concentrations of diesel emissions from fuel and fumes.

## 5 Conclusions

BTEX$_{total}$ concentrations at a diesel refuelling bay in Johannesburg, South Africa, have been noted to have a positive correlation with volume of diesel dispensed daily. Additionally BTEX$_{total}$ indicated to be negatively correlated to temperature changes (thus when atmospheric temperature decreases, BTEX$_{total}$ concentrations increase). This finding is noteworthy, as previous research indicated that total

BTEX concentrations are positively correlated to both volume of fuel (petrol) dispensed and temperature. Benzene concentrations were noted to be neither related to temperature or volume of diesel dispensed, which could be linked to the chemical properties of the benzene vapours. Despite benzene not being significantly related to external factors, p-xylene and ethylbenzene both showed positive correlations to volume of fuel dispensed, while toluene and o-xylene showed a negative relationship with temperature. However, other factors besides total volume of diesel dispensed and temperature (such as wind speed, relative humidity and/or atmospheric pressure) may play a role in the fluctuations of BTEX concentrations, and further evaluation in the future is imperative. In addition, due to the high levels of $\text{BTEX}_{\text{total}}$ concentrations noted at the refuelling bay, it is essential to analyse the health risk exposure of employees in the near future.

## Acknowledgements

The authors would like to thank the National Research Foundation of South Africa for providing funding for this study, and to MetroBus Pty. Ltd. for allowing the research to be conducted on site.

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# CHAPTER THREE: ASSESSMENT OF OCCUPATIONAL EXPOSURE TO BTEX COMPOUNDS AT A BUS DIESEL-REFUELLING BAY: A CASE STUDY IN JOHANNESBURG, SOUTH AFRICA

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**Abstract**

**1. Introduction**

**2. Site Description**

**3. Materials and methods**

**4. Results**

**5. Discussion**

**6. Conclusions and future work**

**Acknowledgements**

**References**

# Assessment of occupational exposure to BTEX compounds at a bus diesel-refueling bay: A case study in Johannesburg, South Africa

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## Abstract

Of increasing concern is pollution by volatile organic compounds, with particular reference to five aromatic hydrocarbons (benzene, toluene, ethyl benzene and two isomeric xylenes; BTEX). These pollutants are classified as hazardous air pollutants. Due to the potential health risks associated with these pollutants, BTEX concentrations were monitored at a bus diesel-refueling bay, in Johannesburg, South Africa, using gas chromatography, coupled with a photo-ionization detector. Results indicate that o-xylene (29-50%) and benzene (13-33%) were found to be the most abundant species of total BTEX at the site. Benzene was within South African occupational limits, but above international occupational exposure limits. On the other hand, occupational concentrations of toluene, ethyl-benzene and xylenes were within national and international occupational limits throughout the monitoring period, based on 8-hour workday weighted averages. Ethyl-benzene and p-xylene concentrations, during winter, correspond to activity at the site, and thus, idling of buses during refuelling may elevate results. Overall, occupational air quality at the refueling bay is a matter of health concern, especially with regards to benzene exposure, and future reduction strategies are crucial. Discrepancies between national and international limit values merit further investigation to determine whether South African guidelines for benzene are sufficiently precautionary.

**Key words:** BTEX; Occupational exposure; Diesel; Occupational exposure limits; Buses

**Highlight:** Occupational concentrations of benzene is considered significantly high (compared to international occupational exposure limits) at a diesel refueling bay in Johannesburg, South Africa, implying that employees could potentially face adverse health effects.

## 1. Introduction

With a global increase in urbanization and number of vehicles and road usage, there has been a concomitant deterioration of air quality in urban areas throughout the world. Thus, in recent years many studies have involved air quality monitoring in cities, pollution hotspot areas, and known hazard risk regions (Brunekreef and Holgate 2002, Fenger 1999, 2009). Air quality effects are particularly significant in developing countries such as South Africa, where rapid urbanization and lack of coherent and integrated transport network planning and management have led to high pollutant emissions (Edokpolo *et al* 2014). Of particular concern from diesel emissions and diesel exhaust fumes are the volatile organic compounds (VOCs). A group of aromatic VOCs, namely BTEX (benzene, toluene, ethyl-benzene and xylenes), includes pollutants known to be harmful to human health, with potential carcinogenic effects (Romieu *et al* 1999, Keretetse *et al* 2008, Tunsaringkarn *et al* 2012, Edokpolo *et al* 2014). This group of VOCs falls within the classification of the World Health Organization’s (WHO) hazardous air pollutants (HAPs) (Table 1). For this reason, monitoring BTEX in the lower atmosphere, specifically in occupational settings where exposure may be greater than in urban areas generally, their effects on both human and ecosystem health has become a priority in recent years (Hoque *et al* 2008, Murena 2007).

**Table 1: Health effects associated with chronic inhalation exposure to BTEX concentrations (Romieu *et al* 1999, Keretetse *et al* 2008, Tunsaringkarn *et al* 2012, World Health Organisation 2012, Moolla *et al* 2013, Edokpolo *et al* 2014)**

| Compound | Health effects from inhalation exposure |
|---|---|
| Benzene | Neurological (central nervous system (CNS) depression: drowsiness, tremors)<br>Respiratory and eye irritant<br>Haematological (blood disorders- aplastic anaemia)<br>Reproductive/developmental (animals: low birth weight, bone marrow damage)<br>Cancer (leukaemia) |
| Ethyl-benzene | Respiratory (throat irritation, chest constriction)<br>Kidney, liver, eye effects<br>Neurological (CNS toxicity) |
| Toluene | Neurological (CNS depression: drowsiness, tremors)<br>Kidney, liver impairment<br>Reproductive/developmental effects |
| Xylenes | Eye, nose, skin, throat irritation<br>Neurological (dizziness, memory loss, headache)<br>Gastrointestinal (nausea, vomiting) |

There are many potential sources of BTEX in air, for example from cigarette smoke, during combustion of gasoline and diesel in motor engines, and petrochemical industries. Research has been conducted on the spatial and temporal variations of VOCs and BTEX in the atmosphere of urban areas, in both developed and developing countries, and shown that BTEX is associated with specific activities such as petrochemical industries, oil refineries or vehicular emissions (Cetin *et al* 2003, Lin *et al* 2004, Lee *et al* 2002). Many studies have focused specifically on petrol refueling stations, gasoline exhaust emissions and health risks related to petrol station workers (for example Das *et al* 1991, Edokpolo *et al* 2014, Hein *et al* 1989, Keretetse *et al* 2008, Oesch *et al* 1995, Onunkwor *et al* 2004, Rekhadevi *et al* 2010, Singh *et al* 2013, Udonwa *et al* 2009). Esteve-Turrillas *et al* (2007) assessed the air quality of BTEX inside vehicles when paused at gasoline filling stations. It was found that risk of exposure to carcinogenic compounds was a calculable risk to human health due to poor insulation or ventilation of vehicle interiors during refueling. In the same manner, it was noted that non-occupationally exposed commuters living and driving in high vehicular traffic areas were also prone to health side effects (such as increased carcinogenic risk). According to Lemire *et al* (2004), there were elevated levels of gasoline components in the blood tests of this sample population in Mexico City. This finding was further reiterated in studies conducted in Iran, by Azari *et al* (2011) and Zoleikha *et al* (2015) where relatively high levels of benzene were found amongst occupationally exposed petrol pump workers.

Benzene levels in gasoline fuel are regulated to 1% content in Australia, Europe and the USA (Caprino and Togna 1998). However, these levels are generally regulated in petrol, while diesel is not a priority. Nelson *et al* (2008) stress that additives to fuels and fuel composition can affect BTEX concentrations and emissions. The differences in practices between developed and developing countries are also amplified, as pump attendants are not as common in developed as they are in developing countries, thus making the issue of occupational health and safety in particular, a developing world issue. As has been noted by Udonwa *et al* (2009), there may be an enhanced level of exposure where pump attendants dispense fuel into the vehicles without using protective devices.

However, there is limited research that examines the effects of diesel exhausts and emissions on air quality (e.g. Carvalho-Oliveira 2005, Fujita *et al* 2011, Heeb *et al* 2000, Tang *et al* 2007, Tsai *et al* 2012) and there are very few studies considering diesel refueling stations and their possible health impacts. In a study conducted on rats exposed to diesel exhaust inhaled chronically at a high concentrations, pulmonary carcinogenicity was noted (Mauderly *et al* 1987). However, none of these studies have focused on BTEX emissions specifically, despite the high toxicity levels of these HAPs. As has been acknowledged by the US Environmental Protection Agency (US EPA), there is growing recognition that diesel fumes and diesel exhaust emissions can have harmful effects on both air quality and human health (Tang *et al* 2007).

Additionally, very little research has been conducted in the Southern hemisphere, and specifically Southern Africa, where vehicular traffic; regulations and compliance; and planning and management may differ from developed countries.

In South Africa, no legislation outlining acceptable levels of VOCs in ambient air exists, making monitoring and emission reduction plans harder to enact (Hoque *et al* 2008). Benzene is the only pollutant that is regularly monitored and is limited in ambient air under legislation by the South African government, despite TEX (toluene, ethyl-benzene and xylenes) also proving harmful to health. This situation however is not unique to South Africa, as there are no standards for TEX in many developed countries either. Nevertheless, there has been a push in South Africa to include limits and guideline values for TEX as they are considered ozone precursor substances (SANS 2011). Moreover, as VOC levels are on the rise (Lee *et al* 2002), the South African government intends to implement a further reduction of benzene limits from 3 ppb to 1.5 ppb by 2016, with no exceedances (above limits) to be allowed (SANS 2011). The primary objective of this study is to evaluate emission characteristics at a diesel bus refueling station in Johannesburg, South Africa. The research includes the following tasks: (a) to collect air concentration data from diesel refueling and exhaust emissions of benzene, toluene, ethyl-benzene, ortho-xylene (o-xylene), and para-xylene (p-xylene); (b) to evaluate whether these chemical concentrations are above legal standards, both national and international; and (c) to investigate the variation of BTEX values during a typical day on site.

## 2. Site Description

The monitoring campaign was carried out at a diesel-refueling bay located in Johannesburg, South Africa (Figure 1). The bus-refueling depot is a government-owned entity that services the public transport sector of the northern suburbs in and around Johannesburg. The refueling bay is under cover with in- and out-flow of fresh air, through four garage doors (which remain open throughout the day and night), 3 m high on either end of the 30 m bay.

There are four refueling pumps, centrally located, where two buses can be refueled simultaneously (Figure 2). The hours of operation are Monday-Friday, 07:30-15:30, and the depot refuels on average 85 buses daily (with on average 6,700 liters pumped per day) during the work hours; with 500 ppm Sulphur diesel. Four full time employees at the depot conduct refueling of the buses on a daily basis.

At the study site, pump attendants wear protective gloves when refueling the buses; however, in South Africa no rubber hood is used over the delivery pump, as is the practice in the USA to reduce emissions (Udonwa *et al* 2009). Additionally, there is very little ventilation, despite bay doors allowing a small amount of airflow to be present, as all four extractor fans located near the ceiling of the bay were non-operational throughout the monitoring period, thus exhaust fumes are not removed rapidly, and may significantly contribute to recorded emissions. Furthermore, 36% of buses at the depot failed the smoke tests conducted by the company. The smoke test indicates whether exhaust fumes released by the bus are within the acceptable range (as determined by the bus manufacturers), as well as if burning of the fuel is occurring correctly. This implies that exhaust fumes within the refueling bay may be further intensified. Spills are also not well managed on site, which further deteriorates the air quality.

This situation however, is not unique to this site. Government-owned public bus companies, throughout South Africa, experience similar conditions. There are 12 urban bus diesel-refuelling stations across the country, all under-cover, and have little to no ventilation and/or filtration systems in place. Additionally, 6 of these refuelling bays service outdated buses that do not pass the smoke tests, but are nevertheless still in operation, due to lack of funds. Moreover, all the government-owned bus depots in the country have full time employed fuel pump attendants. Thus, results and findings from this study can be generalized, and recommendations are applicable in all bus diesel-refuelling stations, and other developing countries, especially in Africa.

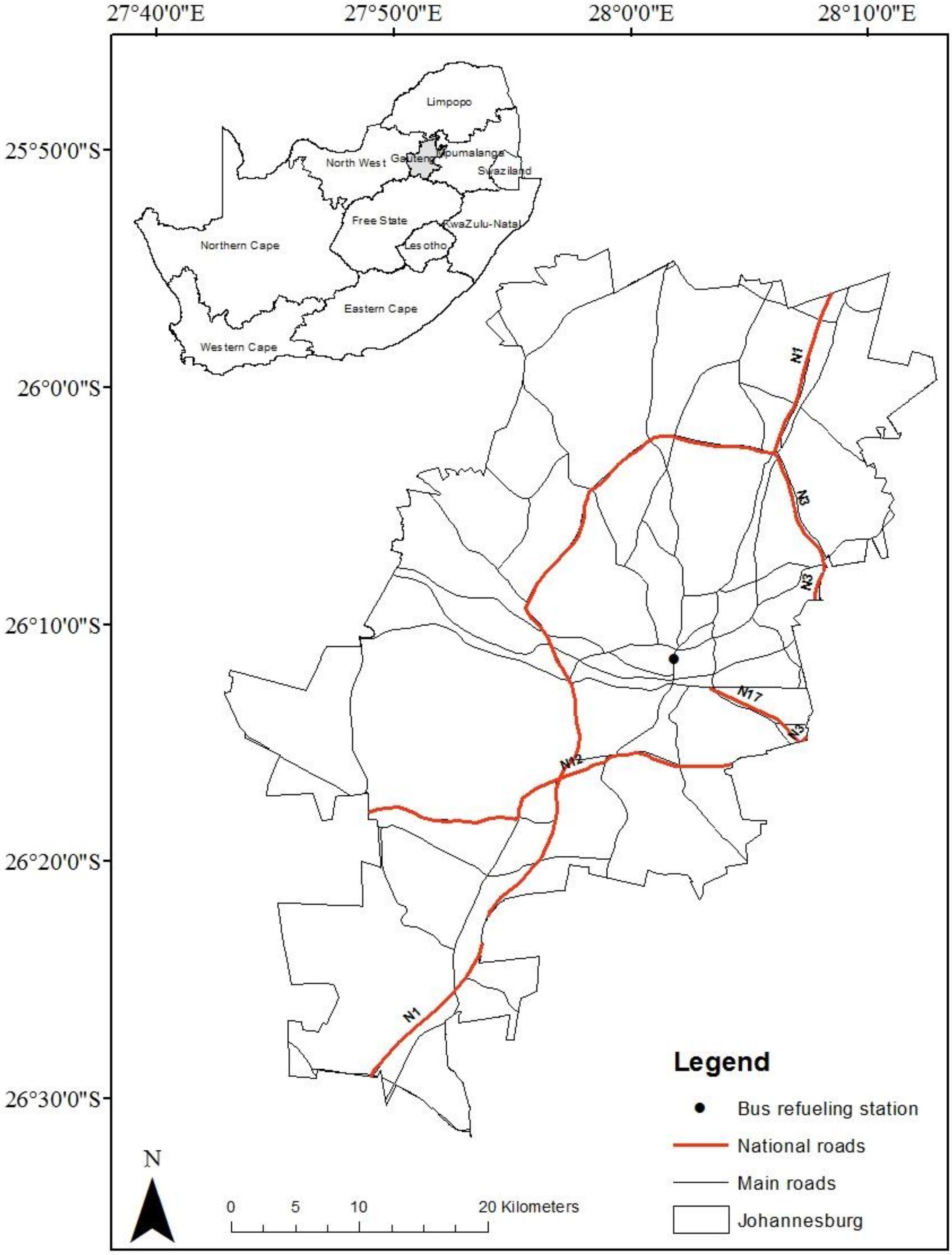

**Figure 1: Map identifying the diesel bus refuelling station situated in Johannesburg CBD, which is in the Gauteng province of South Africa (scale and co-ordinates representative of the Johannesburg region).**

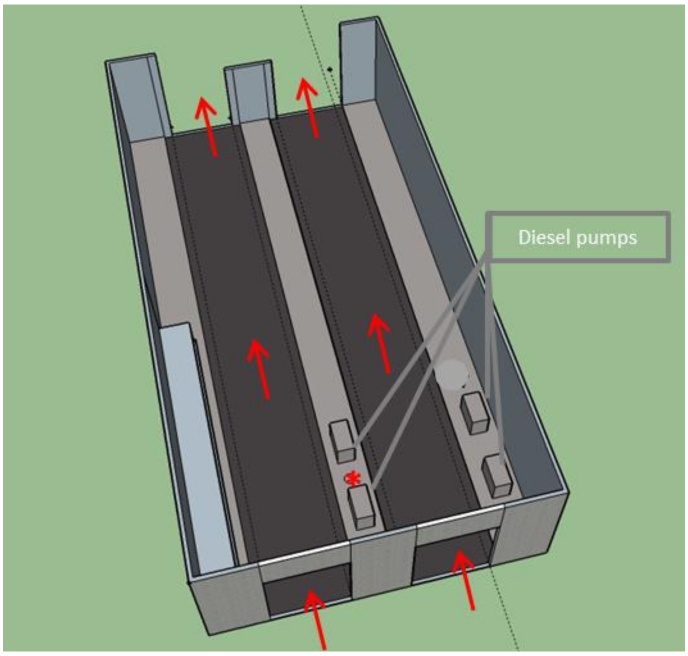

**Figure 2: Diagrammatic representation of the fuel bay on site. The inlet feed for the GC955 instrument (indicated by a red *) was located between the diesel pumps at a height of 1.5 m. Arrows indicate direction of traffic in refuelling bay**

## 3. Materials and methods

Data collection was carried out at an urban diesel-refuelling bay under normal operation conditions throughout the week for a three-month period from June-August 2013 (i.e. winter season). The study period was chosen within the colder, dry season in Johannesburg, when air pollution is commonly higher in the city, and BTEX concentrations are known to be at a maximum (Azari *et al* 2011, Gallego *et al* 2008, Hoque *et al* 2008, Schneider *et al* 2001, Zalel *et al* 2008), which may lead to biases in the data set, i.e. over-estimation compared with annual mean exposure levels. In addition, the short period of monitoring may increase risk of errors. However, a similar study conducted by Truc and Oanh (2007) followed a similar methodology, with the aim to determine exposure levels. Continuous monitoring was carried out for 24 hours, 7 days a week, during both work and non-work hours (13,300 data points in total); to achieve a comprehensive analysis of daily BTEX concentrations, irrespective of activity or inactivity within the fuel bay. The original study design intended to carry out a comparative summer campaign, but high temperatures in the depot caused major instrumental problems. However, data were successfully collected over a 72 hour period in summer, providing some comparative results for another season.

As outlined by Murena (2007), sites that are deemed to have high concentrations of air pollutants can have major implications on human health and, thus, the use of automated systems can be advantageous as continuous data can be collected. For this reason, ambient air was sampled using the Syntech Spectras gas chromatography 955 (GC955), with a photo ionization detector (PID). Additionally, a PID is particularly useful, as it is suited for continuous monitoring and specifically sensitive for aromatic hydrocarbons, even in very low concentrations. The sampling flow was set to sample at 15 minute intervals and sampling height of ambient BTEX was 1.5 m at the center of the fuel bay, with 8 mm tubing and Swagelok fittings used for connecting tubing. Following the methodology of Gallego *et al* (2008) and Shin and Kwon (2000), helium was used as the carrier gas in the GC955, set at a pressure of 350 kPa. The instrument has been tested for accordance to the EMC directive 89/336/EMC, test specification EN 50081-1:1991 and EN 50082-2: 1994. The manufacturer stated error of instrumentation is <2%. A gas mixture (1 ppm of each BTEX) in a gas cylinder (provided by AirLiquide) was adopted to identify elution time and chromatographic peaks. Five aromatic hydrocarbons were continuously monitored (viz. benzene, toluene, ethyl-benzene, and two isomeric xylenes; ortho- and para-xylene). The monitoring instruments were calibrated before use (calibration was done in the range of 0 to 10 ppb). Quality control checks were conducted during and after the monitoring campaign and a correction factor of 2 ppb and 4 ppb for benzene and toluene were used, respectively to counter systematic under-sampling of the instrument. Additionally, 97 data points (<1% of data set) were discarded, as they were distinct outliers.

## 4. Results

The concentrations measured for the winter season of 2013 are shown in Figures 3-4. According to the WHO (World Health Organisation 2012), benzene should not be present in air, as it has been classified as a Group A carcinogen (i.e. known human carcinogen), thus the guideline is set to 0 ppb. Eight hour time-weighted averages (TWA) of TEX concentrations are within South African and US occupational exposure limits (OELs) (Table 2). However, benzene 8-hour TWA concentrations (Figure 3) are above both the United States' National Institute for Occupational Safety and Health (NIOSH) limit and WHO occupational exposure guidelines throughout the monitoring period. This is significant as concentrations are based on operational hours (07:30-15:30), and are 20 times above occupational limits.

**Table 2: Eight hour-time weighted Occupational Exposure Limits (OELs) of NIOSH and South African Occupational Health and Safety (SAOHS) limits, in ppb. 8 hour TWA based on occupational hours (07:30-15:30) (n=4,440) (No OELs available for ethyl-benzene).**

| | Maximum concentration | Minimum concentration | Mean concentration | S.D. | Occupational Exposure Limits | |
|---|---|---|---|---|---|---|
| | | | | | ***SAOHS*** | ***NIOSH*** |
| **Benzene** | 350,00 | 290,00 | 313,16 | 15,18 | 500 | 101 |
| **Toluene** | 450,05 | 38,05 | 188,43 | 110,85 | 46432 | 99498 |
| **Ethyl-benzene** | 639,41 | 0 | 63,72 | 162,56 | n/d | n/d |
| **Xylenes** | 1375,70 | 456,63 | 850,97 | 286,63 | 100186 | 100186 |

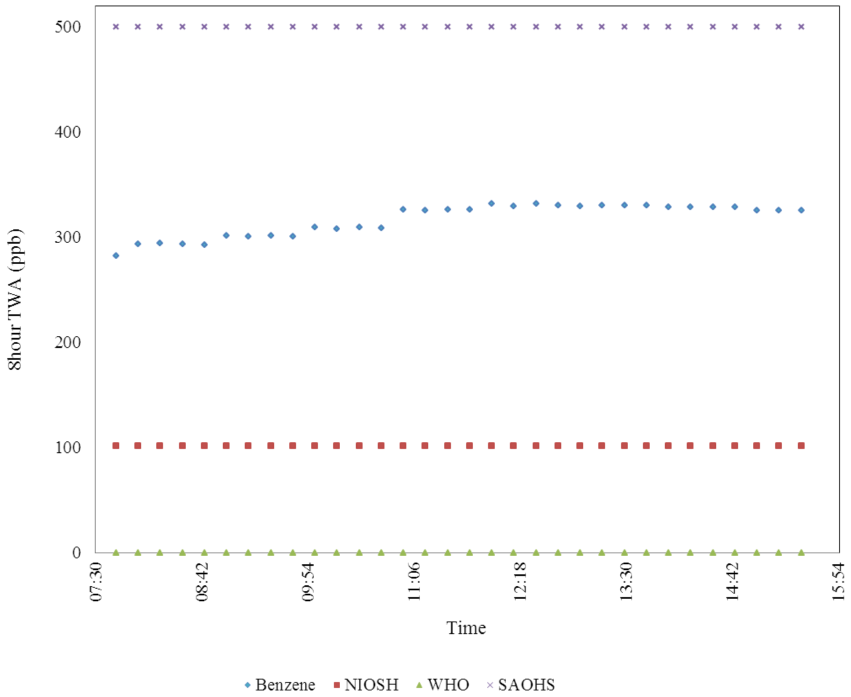

**Figure 3: Benzene 8hour time weighted average (TWA) concentrations for the winter monitoring period of 2013, indicating that concentrations during occupational hours (07:30-15:30) were significantly higher than the World Health Organization's (WHO) guidelines and National Institute for Occupational Safety and Health (NIOSH) limits throughout the monitoring period, but are within South African Occupational Health and Safety (SAOHS) national standards.**

As can be noted in Figure 4, there is a distinct pattern with regards to the BTEX concentrations in both summer and winter. However, due to the fact that the instrument malfunctioned in hotter temperatures (thus a lack of data capturing during 09:00-12:00), some findings are inconclusive. Additionally, the summer monitoring campaign only consisted of 72 hours of data (1,080 data points), and thus may not be fully indicative of the pattern throughout the season. Nevertheless, $BTEX_{total}$ (where $BTEX_{total}$ is a cumulative amount of individual BTEX species), is significantly higher in the winter season, even in the southern hemisphere, where synoptic climate conditions may differ.

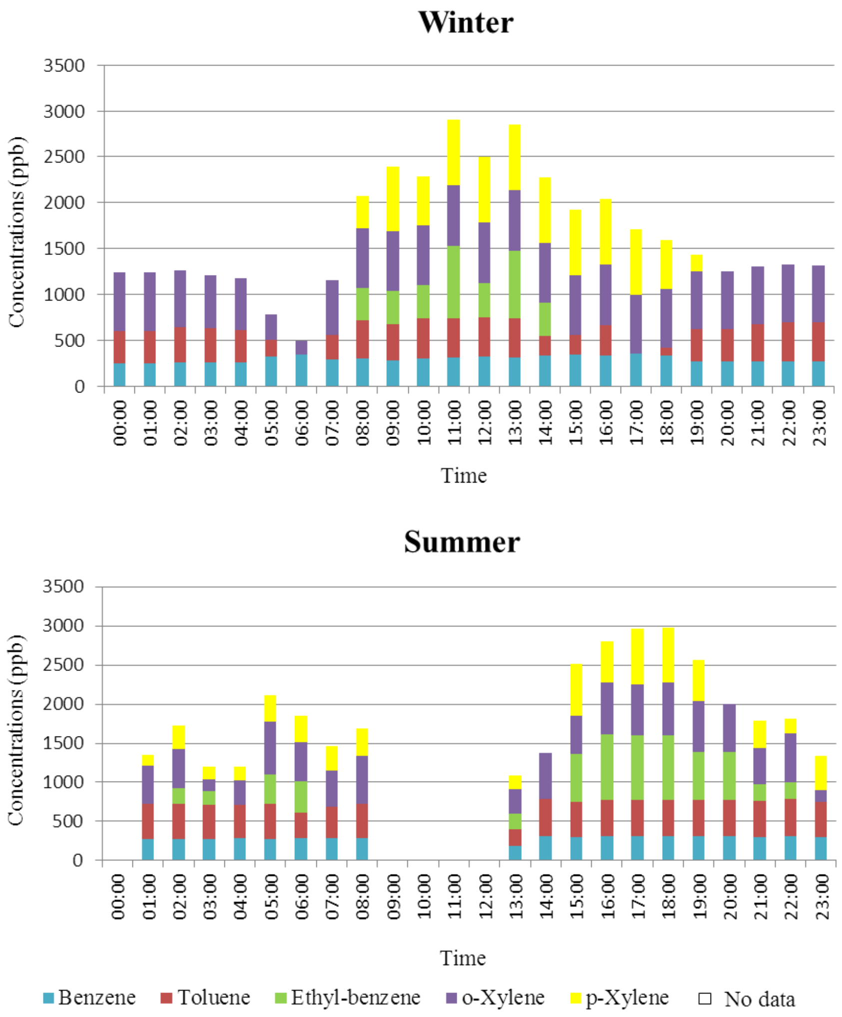

Figure 4: A typical day, indicating levels of BTEX species, in winter (13 July 2013) and summer (02 January 2014) conditions.

## 5. Discussion

Benzene is naturally ubiquitous in the atmosphere, however, high levels of exposure to benzene is considered a risk to the health of workers. Thus, at this site, a matter of concern is that benzene concentrations are above both the NIOSH and WHO occupational exposure guidelines (Figure 3) which can lead to adverse health effects of employees at the site, as many studies have argued that long term inhalation of benzene increases carcinogenic or mutagenic incidences (Perry and Gee 1995). However, results are still within South African Occupational Health and Safety (SAOHS) standards. It is also noteworthy to mention the significant difference in international and national OELs reflected in Table 2, of benzene, toluene and xylenes. International limits for benzene are significantly lower than South African national standards, while the reverse is true for toluene limits. This in itself is a point of concern as benzene is a carcinogen, even at low level exposure.

There are some interesting features of the current dataset which are worth emphasising. In general, it is expected that emissions should increase throughout the day as refuelling continues. In winter, the pattern noted at this site relates to the refuelling pattern of the buses at the site. Ethyl-benzene and p-xylene concentrations occurred only from 08:00; 30 minutes after refuelling begins at the site, and occurs mainly during refuelling times (i.e. opening times). Similar trends were noted by Zalel *et al* (2008), where specific concentrations increase during refuelling.

Changes in levels of BTEX can be attributed to traffic movement, with buses frequently idling during refueling, as well as vapours from refueling itself. In Antwerp, Belgium, where road reconstruction projects took place to reduce air pollution by reducing the number of lanes for vehicular traffic on a high traffic route, it was noted that air quality deteriorated (Buczynska *et al* 2009). This deterioration was attributed to intensified traffic and ‘stand-still traffic’, thus exhaust emissions increased, placing pressure on air quality limits. In other studies, it was found that there were high concentrations of BTEX inside air environments of passenger buses in Changsha, China, and that toluene and xylene were also above local indoor air quality standards (Chen *et al* 2011). However, exposure time was shorter, as commuters exited the buses on reaching their destination. Elsewhere in China, Yu and Li (2014) found that many different factors affected air quality near bus stops; including distance to road-side, height of curb, and traffic intensity; where diesel fueled public buses were located; however, the focus of this study was not on BTEX emissions.

Despite benzene comprising at most one third of the BTEX species (o-xylene was found to be the most abundant species varying from 29-50% of $BTEX_{total}$ at the site, while benzene comprised 13-33%), with similar patterns observed in a study by Hoque *et al* (2008), benzene is a matter of concern due its worldwide distribution and known carcinogenic effects, even in lower concentrations (Duarte-Davidson *et al* 2001, World Health Organisation 2012). It has a relatively long life span, low reactivity and is stable in the atmosphere. However, during summer months, where benzene concentrations are observed to be lower than in winter, benzene undergoes chemical degradation (Gallego *et al* 2008, Hoque *et al* 2008, Schneider *et al* 2001, Zalel *et al* 2008). Benzene also evaporates rapidly at room temperature, is highly flammable and can be inhaled, or ingested through the skin easily on contact. The fact that benzene levels are significantly high at the site should indicate the need for continuous monitoring and regulatory systems in place.

On the other hand, toluene and xylenes are shown to evaporate in hotter climates, decreasing ground level concentrations (GLCs) (Hoque *et al* 2008). Whilst it is understood that toluene and xylenes have a lower toxicity level than that of benzene, when exposed to photochemical reactions in the atmosphere they too can react to form new compounds which can result in adverse health effects (Gallego *et al* 2008).

## 6. Conclusions and future work

This paper demonstrates that of the five aromatic hydrocarbons analyzed, only benzene exceeded international occupational limits, but by as much as a factor of 20, throughout the winter monitoring period. Such exceedances of allowed exposure limits in the workplace imply many potential long-term health effects potentially include cancers, neurological, respiratory, hematological and/or reproductive disorders, for employees at the site.

Furthermore, at retail gasoline stations in South Africa, where attendants refuel motor vehicles, trucks and buses with both petrol and diesel on a daily basis, no protective gear is worn by attendants, nor are the stations "self-service" as is the case in many developed countries. This has implications for retail stations, where no studies have examined the effects on diesel pump attendants, despite being exposed to these vapours and fumes on a daily basis. Thus, research on this topic is not only important, but wider studies are imperative to analyze potential environmental exposure risk directly to employees.

At sites such as these where occupational limits are highly exceeded, it is imperative for ventilation and filtration systems to be put into place and maintained. Other factors which exacerbate air quality problems should also be urgently addressed. BTEX concentrations may be elevated by fuel spillages or idling of buses, releasing exhaust fumes, during refueling. Protocols for dealing with accidental spillages and unnecessary exposure to emissions should be implemented in such situations as a matter of priority.

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# CHAPTER FOUR: OCCUPATIONAL EXPOSURE OF DIESEL STATION WORKERS TO BTEX COMPOUNDS AT A BUS DEPOT

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**Abstract**

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*Article*

# Occupational Exposure of Diesel Station Workers to BTEX Compounds at a Bus Depot

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**Abstract:** Diesel fuel is known to emit pollutants that have a negative impact on environmental and human health. In developing countries like South Africa, attendants are employed to pump fuel for customers at service stations. Attendants refuel vehicles with various octane unleaded fuel, lead-replacement petrol and diesel fuel, on a daily basis. Attendants are at risk to adverse health effects associated with the inhalation of volatile organic compounds released from these fuels. The pollutants released include benzene, toluene, ethylbenzene and xylenes (BTEX), which are significant due to their high level of toxicity. In this study, a risk assessment of BTEX was conducted at a diesel service station for public buses. Using Radiello passive samplers, it was found that benzene concentrations were above recommended international standards. Due to poor ventilation and high exposure duration, the average benzene concentration over the sampling campaign exceeded the US Environmental Protection Agency's chronic inhalation exposure reference concentration. Lifetime cancer risk estimation showed that on average there is a $3.78 \times 10^{-4}$ cancer risk, corresponding to an average chronic daily intake of $1.38 \times 10^{-3}$ mg/kg/day of benzene exposure. Additionally, there were incidences where individuals were at potential hazard risk of benzene and toluene that may pose non-carcinogenic effects to employees.

**Keywords:** diesel; BTEX; health risk assessment; lifetime cancer risk; hazard quotient

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## 1. Introduction

Inhalation of pollutants such as volatile organic compounds (VOCs) has been shown to have many side-effects on human health. A group referred to as BTEX (benzene, toluene, ethylbenzene and the three isomers of xylene) has been found to be potentially hazardous to environmental and human health [1]. Human exposure to BTEX, both through inhalation or ingestion, can have serious health impacts, such as neurological diseases, cancers, and teratogenic effects [2–4]. This is of major concern, as Chauhan *et al.* [2] state that 50% of BTEX inhaled by humans over a person's lifespan is actually absorbed into the body. The World Health Organization (WHO) estimate that 4 in 1 million people are at risk of developing leukemia in their lifetime when exposed to 1 mg/m$^3$ of benzene [5].

In South Africa, as is the case in many developing countries, people are still employed to refuel vehicles, trucks and buses at gas stations. In South Africa petrol pump attendants refuel vehicles with lead replacement petrol (LRP); 93-unleaded and 95-unleaded petrol; and 200, 50, or 10 ppm sulphur diesel, on a daily basis [6]. As such, attendants are particularly at risk to adverse health effects associated with inhalation of hazardous air pollutants (HAPs), such as BTEX which are released from these fuels. These attendants are thus exposed to both petrol and diesel fumes daily. However, despite numerous studies investigating the effects of gasoline inhalation on petrol pump workers and auto mechanics, there are no health risk assessment (HRA) studies focusing on diesel pump workers, despite most retail garages in developing countries providing both petrol and diesel services (Table 1). The purpose of this paper is to perform a site-specific health risk analysis to investigate the occupational exposure to BTEX, and inhalation risk, of workers at a diesel refueling station in South Africa. As many auto mechanics also face the risk of adverse health effects when exposed to BTEX, the health analysis will focus on both diesel pump attendants, as well as auto mechanics at a bus depot.

**Table 1.** A review of health risk assessments (HRAs), of various volatile organic compounds (VOC), specifically benzene, toluene, ethyl-benzene and xylenes (BTEX) studies, and conducted at/near petrol (gasoline) filling stations, in chronological date order. (BTX—Benzene, toluene and xylenes).

| Location | Focus Area | Sampling Method | Ref. |
|---|---|---|---|
| Rangoon, Burma | Occupational benzene exposure in petrol filling stations | Urine samples | [7] |
| Kanpur/Lucknow, India | Environmental impact on health of workers at retail petrol pumps | Rotheroe and Mitchell personal samplers | [8] |
| Mexico City | Environmental exposure to VOCs among workers | Passive organic vapour badges and blood samples | [9] |
| Prunay, France | BTX concentrations near a stage II implemented petrol station | Gas chromatography + flame ionisation detector | [10] |
| -- | Occupational exposure to benzene in gasoline filling station attendants | Radiello passive samplers and urine samples | [11] |
| Valencia, Spain | Air quality of BTEX inside vehicles and at gasoline filing stations | semipermeable membrane devices | [3] |
| Ioannina, Greece | Ambient benzene concentrations in the vicinity of petrol stations and associated health risk | Passive and active samplers | [12] |

**Table 1.** *Cont.*

| Location | Focus Area | Sampling Method | Ref. |
|---|---|---|---|
| Ioannina, Greece | Assessment and prediction of exposure to benzene of filling station employees | Active and passive samplers | [13] |
| Chonburi, Thailand | HRA of VOCs in gas service station workers | Urine samples and air samplers | [14] |
| Kolkata, India | VOCs at petrol pumps: Exposure of workers and HRA | Personal air samplers | [15] |
| Calaba, Nigeria | Exposure of petrol station attendants and auto mechanics to petrol fumes | Structured questionnaires, venous blood samples analysis | [6] |
| Hyderabad, India | Geno-toxicity of filling station attendants exposed to petroleum hydrocarbons | Blood samples and Comet Assay | [16] |
| Murcia, Spain | Assessing the impact of petrol stations on their immediate surroundings | Radiello passive samplers | [17] |
| Montreal, Canada | BTEX exposures in automobile mechanics and health risks | Active chemical ionisation mass spectrometry | [18] |
| Bangkok, Thailand | Occupational exposure of gasoline station workers to BTEX compounds | Active samplers | [19] |
| India | Occupational health exposure at petroleum refinery | Organic vapour samplers | [20] |
| Bangkok, Thailand | HRA of petrol station workers and assessing exposure of inhaling BTEX | Personal air samplers | [21] |
| Multiple areas | HRA of BTX in gasoline service stations | BTX exposure data from scientific literature | [22] |
| Australia | Leukaemia and exposure to benzene in petroleum workers | Diagnostic information | [23] |
| Johannesburg, South Africa | Air quality of BTEX at a diesel filing station | Gas chromatography + photo ionisation detector | [24] |

As a case study, this work is applied to a bus depot where there is significant movement of diesel buses. This analysis can thus provide useful information about potential health risks associated with BTEX vapours released from diesel refueling pumps and exhaust emissions.

## 2. Experimental Section

### 2.1. Study Site

The monitoring campaign was carried out at a bus depot located in central Johannesburg, South Africa. A government owned entity, which supplies the public transportation routes in central and northern Johannesburg, manages the depot. The bus depot accommodates 400 buses, where refueling, repairs and general maintenance of the buses are handled. The operating hours of the refueling bay and adjacent workshop is from 07:30 to 15:30, Monday–Friday.

The buses are fueled with standard 500 ppm diesel, in the refueling bay on site. The refueling bay consists of four diesel pumps, with four full time employed personnel. The refueling bay is undercover, with large 3 m high doors on either end of the bay. Bus engines continue running while refueling, thus exhaust fumes are present in the bay as well as vapors from refueling. In close proximity to the refueling bay (Figure 1a) is a large enclosed workshop where maintenance and repairs take place (Figure 1b). There is very little ventilation in the workshop, and all filters and extraction fans on site are out of order in both workspaces.

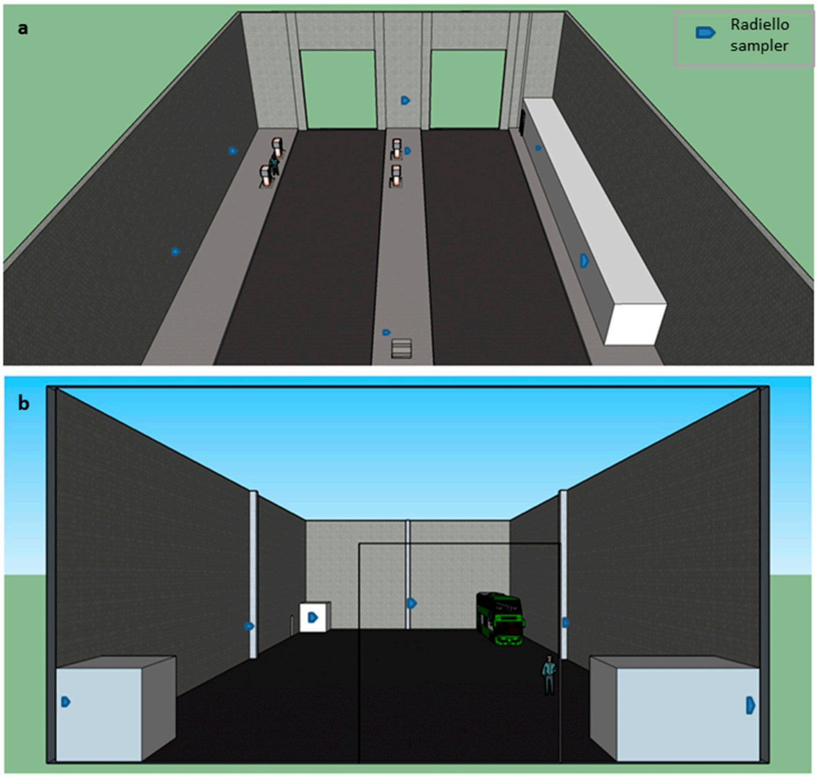

**Figure 1.** Radiello passive sampler positions in the (**a**) fuel bay and (**b**) workshop. Passive samplers were positioned equidistant as possible, at 2 m heights. (Overhead roofs and front walls have been made transparent for viewing purposes. Offices are displayed as white boxes). The fuel bay is adjacent to the workshop at the bus depot.

### *2.2. Study Sample*

The occupationally exposed group consisted of fuel bay attendants (FBA) from the refueling bay ($n$ = 4) and diesel auto mechanics (AM) from the bus workshop ($n$ = 16). Only full time employees were considered for this study. The demographic information provided by the bus company is illustrated in Table 2. All employees worked a standard 8 hours per day, 5 days a week.

The majority of the employees at the site were male (only one auto-mechanic at the workshop was female). Overall, the majority of the employees did not smoke (75%). As can be seen in Table 2 there is a very wide range of employee ages (24 to 63 years) and exposure duration (from 1 to 41 years).

**Table 2.** Demographic data of participants in the fuel bay attendants (FBA) and auto-mechanics (AM). Data supplied by bus operating company.

| Participant ID | Workplace | Gender | Smoker | Age (years) | Employment Duration (years) |
|---|---|---|---|---|---|
| FBA1 | Fuel Bay | Male | No | 27 | 5 |
| FBA2 | Fuel Bay | Male | No | 45 | 10 |
| FBA3 | Fuel Bay | Male | No | 59 | 33 |
| FBA4 | Fuel Bay | Male | Yes | 56 | 37 |
| AM1 | Workshop | Male | Yes | 27 | 1 |
| AM2 | Workshop | Male | Yes | 35 | 1 |

**Table 2.** *Cont.*

| **Participant ID** | **Workplace** | **Gender** | **Smoker** | **Age (years)** | **Employment Duration (years)** |
|---|---|---|---|---|---|
| AM3 | Workshop | Male | No | 25 | 2 |
| AM4 | Workshop | Male | No | 26 | 2 |
| AM5 | Workshop | Male | No | 24 | 2 |
| AM6 | Workshop | Male | No | 25 | 3 |
| AM7 | Workshop | Male | No | 29 | 4 |
| AM8 | Workshop | Male | No | 36 | 5 |
| AM9 | Workshop * | Female | No | 47 | 8 |
| AM10 | Workshop * | Male | Yes | 40 | 10 |
| AM11 | Workshop | Male | No | 41 | 10 |
| AM12 | Workshop | Male | Yes | 51 | 11 |
| AM13 | Workshop | Male | No | 40 | 16 |
| AM14 | Workshop | Male | No | 38 | 16 |
| AM15 | Workshop | Male | No | 49 | 28 |
| AM16 | Workshop * | Male | No | 63 | 41 |

* Employee based within an enclosed office inside the workshop.

### *2.3. Passive Sampling Strategy*

Sampling and monitoring occurred during the winter, as many studies using both active and passive sampling strategies have established that BTEX concentrations are elevated in winter as compared to other seasons [4,25–27]. Johannesburg, located on the interior plateau of the country, experiences cold, dry winters, with temperatures ranging from −3 to 19 °C. A Luft Weather Sensor, with built-in temperature, humidity, wind speed and wind direction sensors, was deployed within the refueling bay, as there are garage doors that can allow for in/out flow of fresh air.

Radiello passive air samplers were arranged in both the refueling bay and workshop (Figure 1), where emissions were considered to be at their maximum. These samplers are considered reliable in both indoor and outdoor environments, and following the European standard (EN 13528-2), the Radiello passive samplers were used to analyze the risk exposure of BTEX in this situation [28]. Concentrations as low as 2 $\mu g \cdot m^{-3}$ may be measured with the samplers, with an error not exceeding 0.1 $\mu g \cdot m^{-3}$. The samplers were deployed for 14 days as prescribed by the manufacturer. The BTEX passive samplers consist of an absorbing cartridge, which is placed in a micro-porous polyethylene membrane surface (50 mm long micro-porous cylinder; 16 mm external diameter; 300 mg of 40–60 mesh Carborograph 4) [28,29]. Each cartridge was secured to a triangular polycarbonate supporting plate. The sampling plates and cartridges were not placed within protective chambers, as wind speeds were low enough to avoid error [30] (average wind speeds were 2 $m \cdot s^{-1}$ in the refueling bay and workshop, as they are undercover sites). As outlined by Gallego *et al.* [25], each sample was labelled, and initial and final sampling times were recorded. As soon as the sampling process was over, tubes were returned to their protective containers and sent to a laboratory for analysis (ChemTech Labs, Johannesburg, South Africa). Tubes containing the samples were stored in a dark, cool box. As advised by the laboratory, leaded pencils were avoided as to preclude any contamination of the samples.

### *2.4. Risk Characterization*

As has been shown in many studies, inhalation risk analysis is vital in order to determine the potential exposure of employees [15,19,21,31]. Both cancer risk and hazard risk calculations (associated with the inhalation of air pollutants) were done for employees to evaluate the potential effect of BTEX on human health. Individual calculated cancer risk and hazard risk values were compared with the United States Environmental Protection Agency (US EPA) acceptable standards.

To calculate cancer risk (CR) Equation (1) was applied, while Equation (2) was used to evaluate the non-carcinogenic hazard quotient (HQ):

$$Cancer\ Risk\ (CR) = Lifetime\ Average\ Daily\ Dose\ (LADD) \times Slope\ Factor \tag{1}$$

$$Hazard\ Quotient\ (HQ) = Lifetime\ Average\ Daily\ Dose\ (LADD)/reference\ dose \tag{2}$$

Inhalation slope factor (SF) [benzene 0,0273 $(\mathrm{mg/kg/day})^{-1}$] and reference dose (RfD) standard values were used [benzene 0,00855 $\mathrm{mg \cdot kg^{-1} \cdot day^{-1}}$, toluene 1.43 $\mathrm{mg \cdot kg^{-1} \cdot day^{-1}}$ and xylenes 0,029 $\mathrm{mg \cdot kg^{-1} \cdot day^{-1}}$] [22,31].

To calculate the Lifetime Average Daily Dose (LADD) of employees, Equation (3) was utilised:

$$LADD = (C.CF.IR.EF.ED)/(BW.AT) \tag{3}$$

where C is the contaminant concentration (average concentrations used from passive samplers) ($\mu\mathrm{g/m^3}$); CF is the conversion factor (1 mg/1000 μg); IR is the inhalation rate (US EPA standard) (20 $\mathrm{m^3}$/day); EF is the exposure frequency (days/year); ED is the exposure duration (years); BW is body weight (US EPA standard) (70 kg); and AT is the averaging time (exposure averaged over life time/average life expectancy for male and female) (days).

Demographic data were provided by the operations manager, in order to provide information pertaining to individual input variables such as age, sex, exposure frequency and exposure duration (Table 2). Where data were limited, US EPA standard values were used for body weight (70 kg for males and 60 kg for females) and inhalation rate (20 $\mathrm{m^3}$/day) [31]. In order to calculate the exposure frequency, standard values were used (all employees worked a standard five days, eight hours per day and received a minimum of 21 days leave per annum). More detailed evaluation of confounding factors, such as smoking habits and home conditions, was beyond the scope of this study.

## 3. Results

### *3.1. BTEX Monitoring*

BTEX concentrations from passive samplers are shown in Table 3. Averages of these concentrations have been used to analyse the potential risk of employees (*i.e.*, contaminant concentration (Equation (3)). The average benzene ambient concentration results in the general fuel bay and workshop areas as well as the workshop offices (Table 3) pose a potential cancer risk for employees, as the World Health Organisation (WHO) states that benzene is a known human carcinogen and thus no safe level of exposure can be recommended. Regarding the workshop, there is a statistically insignificant difference between concentrations found within individual offices and the general area of the workshop; however, concentrations are slightly lower in the offices. One important factor to note is the higher levels of toluene and xylenes in the general area of the workshop (maximum concentrations of 11.93 and 13.12 ppb, respectively), as compared to all other workspaces at the study site.

**Table 3.** Average BTEX concentrations (in ppb) from Radiello passive samplers in the refueling bay and workshop (average atmospheric temperature during the monitoring period was 14.3 °C; six samplers were placed in each workspace).

| | **Benzene** | **Toluene** | **Ethylbenzene** | **Xylenes** |
|---|---|---|---|---|
| **Fuel Bay General Area** | | | | |
| Geometric Mean | 1.21 | 2.26 | 0.57 | 3.52 |
| Max | 1.26 | 2.43 | 0.87 | 4.97 |
| Min | 1.16 | 2.13 | 0.42 | 2.09 |
| s.d | 0.15 | 0.57 | 1.06 | 2.72 |
| **Workshop—General Area** | | | | |
| Geometric Mean | 1.41 | 3.22 | 0.64 | 3.97 |
| Max | 1.65 | 11.93 | 3.35 | 13.12 |
| Min | 1.25 | 2.33 | 0.41 | 2.25 |
| s.d | 0.66 | 9.82 | 7.30 | 10.03 |
| **Workshop—Offices** | | | | |
| Geometric Mean | 1.38 | 2.76 | 0.67 | 4.10 |
| Max | 1.48 | 3.00 | 0.96 | 4.79 |
| Min | 1.29 | 2.46 | 0.50 | 2.84 |
| s.d | 0.32 | 1.08 | 1.05 | 4.40 |

### *3.2. Health Risk Assessment*

According to the US EPA, a cancer risk above $1 \times 10^{-6}$ is unfavourable, as it significantly increases carcinogenic potential in humans. All employees exceed the critical guideline value at this study site (Table 4). Of particular concern are participants FBA4 and AM16 (Table 4), as they have a potential risk of 1 in 1000 chance of developing cancer ($1 \times 10^{-3}$). These two employees have been employed the longest, at 37 and 41 years, respectively.

**Table 4.** Lifetime potential cancer risk for individual participants from exposure to benzene. The potential risk of $1 \times 10^{-5}$ = 1 in 100,000; $1 \times 10^{-4}$ = 1 in 10,000; and $1 \times 10^{-3}$ = 1 in 1000 is based on the probability of developing cancer in a population sample.

| **Participant ID** | **Cancer Risk** |
|---|---|
| FBA1 | $1.37 \times 10^{-4}$ |
| FBA2 | $2.74 \times 10^{-4}$ |
| FBA3 | $9.03 \times 10^{-4}$ |
| FBA4 | $1.01 \times 10^{-3}$ |
| AM1 | $3.24 \times 10^{-5}$ |
| AM2 | $3.24 \times 10^{-5}$ |
| AM3 | $6.47 \times 10^{-5}$ |
| AM4 | $6.47 \times 10^{-5}$ |
| AM5 | $6.47 \times 10^{-5}$ |
| AM6 | $9.71 \times 10^{-5}$ |

**Table 4.** *Cont.*

| Participant ID | Cancer Risk |
|---|---|
| AM7 | $1.29 \times 10^{-4}$ |
| AM8 | $1.62 \times 10^{-4}$ |
| AM9 | $2.72 \times 10^{-4}$ |
| AM10 | $3.07 \times 10^{-4}$ |
| AM11 | $3.24 \times 10^{-4}$ |
| AM12 | $3.56 \times 10^{-4}$ |
| AM13 | $5.18 \times 10^{-4}$ |
| AM14 | $5.18 \times 10^{-4}$ |
| AM15 | $9.06 \times 10^{-4}$ |
| AM16 | $1.39 \times 10^{-3}$ |

A hazard quotient (HQ) is a measure of potential overall hazard risk. A HQ of ≥1 is considered as an "adverse non-carcinogenic effect of concern"; while a value of <1 considered an "acceptable level" [19]. Thus, xylene concentrations at the site pose a low potential hazard risk, and are within acceptable standards (Table 5). However, benzene and toluene HQ are high, implying potential adverse health effects to employees.

**Table 5.** The hazard quotient (HQ) for benzene, toluene and xylenes; indicating the potential hazard risk to employees on exposure to compounds. An HQ >1 is considered an adverse non-carcinogenic effect of concern. HQ levels ≥1 are in bold for individual participants.

| Participant ID | Hazard Quotient | | |
|---|---|---|---|
| | Benzene | Toluene | Xylenes |
| FBA1 | 0.717 | **1.588** | 0.046 |
| FBA2 | **1.433** | **3.176** | 0.092 |
| FBA3 | **4.731** | **10.480** | 0.305 |
| FBA4 | **5.304** | **11.751** | 0.342 |
| AM1 | 0.170 | 0.452 | 0.012 |
| AM2 | 0.170 | 0.452 | 0.012 |
| AM3 | 0.339 | 0.904 | 0.024 |
| AM4 | 0.339 | 0.904 | 0.024 |
| AM5 | 0.339 | 0.904 | 0.024 |
| AM6 | 0.509 | **1.356** | 0.036 |
| AM7 | 0.678 | **1.808** | 0.048 |
| AM8 | 0.848 | **2.260** | 0.060 |
| AM9 | **1.424** | **3.559** | 0.115 |
| AM10 | **1.610** | **3.848** | 0.124 |
| AM11 | **1.695** | **4.521** | 0.120 |
| AM12 | **1.865** | **4.973** | 0.132 |
| AM13 | **2.712** | **7.233** | 0.192 |
| AM14 | **2.712** | **7.233** | 0.192 |
| AM15 | **4.746** | **12.659** | 0.335 |
| AM16 | **7.261** | **15.778** | 0.509 |

Employees that have worked for more than 30 years are especially at risk to adverse non-carcinogenic effects (*i.e.*, FBA3-4 and AM15-16). This is further illustrated in Figure 2, where it is evident that with increasing work duration, there is a significant increase in both potential cancer and hazard risks. In addition to exposure duration playing a role in potential risk, placement within the workshop also plays a role. Thus, the range of risk is large at times, where inhalation exposures differ.

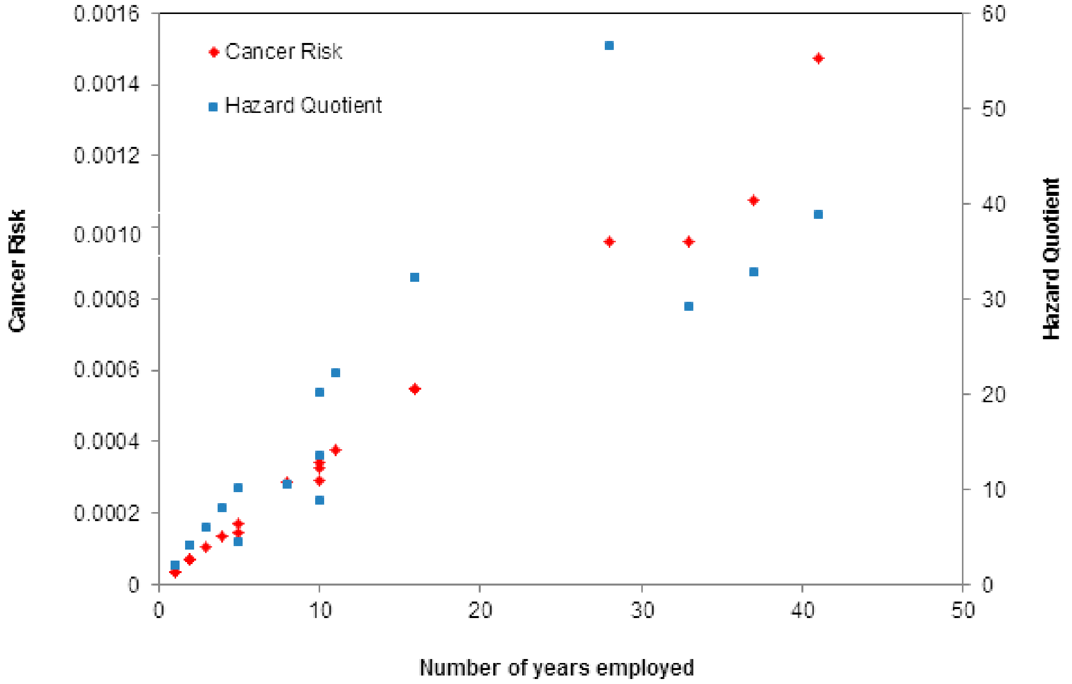

**Figure 2.** Cancer risk and hazard quotient (combined benzene, toluene and xylene hazard quotients) as compared to number of years employed.

## 4. Discussion

### 4.1. BTEX Monitoring

The average benzene concentrations from the passive samplers measured at this site (Table 3) are significantly higher than the average concentrations of benzene measured in the atmospheric air of many European cities (e.g., Belgium, Greece, Italy, *etc.*). However, concentrations from the European studies are annual average concentrations, where concentrations may be decreased in summer months. Higher concentrations of benzene are generally reported in winter months, as a lower average ambient temperature has been noted to contribute to the accumulation of pollutants in the atmosphere as there is reduced movement of air masses in the upper atmosphere [29]. This study was conducted during the winter season, and consequently, results may be elevated relative to annual means. Furthermore, the ambient measurements from the European cities may not be site specific, compared to refuelling bays and workshops where levels may be intensified, as they may be generalised ambient air quality monitoring campaigns. However, mean benzene concentration in the fuel bay and workshop general areas, as well as the workshop offices (1.20; 1.40; 1.37 ppb, respectively) exceeds the US EPA's inhalation exposure reference concentration (1.2 ppb) and the chronic inhalation reference concentration ($3 \times 10^{-5}$ ppb), as well as the WHO air quality guideline (0 ppb).

When buses are re-fuelled, diesel vaporisation and diesel exhaust emissions from the idling buses contribute to the increased benzene concentrations. The fuel bay serves as a source of diesel fuel in this study, however average benzene emissions are higher in the workshop than the fuel bay. There is not sufficient evidence to explain this relationship, but it does reveal the importance of diesel exhaust emissions when considering human health in occupational environments, as ventilation and extraction mechanisms are not in place at the study site. It should be noted that the workshop is generally more active with idling buses being repaired by auto-mechanics throughout the day, with spray painting activities also occurring, which may increase BTEX concentrations.

Although adjustments have been made to reduce the percentage volume of benzene in diesel fuel there is still a global trend of increased benzene emissions near fuel stations [19,24,29,32]. Edokpolo *et al.* [22] postulate that vaporisation inside fuel stations is the main sources of benzene in the atmosphere nearby. In a study conducted by Karakitsios *et al.* in Greece, similar studies indicated that even in developed countries where vapour recovery systems exist, filling station attendants are still exposed to high benzene concentrations (5–16 ppb) [13]. Benzene levels were also found to be directly proportional to volume of fuel dispensed [12,13].

### *4.2. Quantitative Risk Analysis*

Results from the lifetime cancer risk estimation on employees at the bus depot show that on average there is a $3.78 \times 10^{-4}$ cancer risk, corresponding to an average chronic daily intake of $1.38 \times 10^{-3}$ mg·kg$^{-1}$·day$^{-1}$ of benzene exposure. This implies that, on average, there is 3-in-10,000 chance that employees at the site may develop cancer in their lifetime, with some employees experiencing even higher probabilities (Table 4). The lifetime cancer risk thus exceeds the US EPA standard of $1 \times 10^{-6}$ for all employees (Table 4). Health risk assessments conducted in a wide variety of environments reiterate this finding. Studies indicate that cancer risks of sample groups exposed to benzene concentrations generally exceed the US EPA cancer risk limit [32–37].

It was determined by Guo *et al.* [38] that inhalation exposure to benzene accounts for more than 40% of cancer risks for various indoor environments. This finding was confirmed in other studies where benzene baseline blood levels were higher in groups exposed to constant BTEX emissions, compared to those that are not exposed. It was found that benzene levels in blood were directly proportional to benzene concentrations in the atmosphere, specifically in fuel stations [9,33]. Romieu *et al.* [9] also determined that the blood baseline benzene levels of fuel attendants did not increase over the work shift as expected. This was attributed to chronic level of exposure, and not short term exposure, thus determining potential lifetime cancer risks is essential in high risk areas.

At this study site, the majority of the employees did not smoke, however many studies have confirmed that long term exposure to volatile organic compounds from diesel exhaust emissions increased the cancer risk among smokers and non-smokers alike [39]. Weisel [40] found that benzene inhalation exposure in occupational settings may be increased in employees who smoke. However, Oesch *et al.* [41] suggested that smokers are sporadically less affected to BTEX inhalation as smoking has a detoxifying effect. Regrettably, this study could not take into account the exact effect of smoking when exposed to high levels of BTEX, and analyze the potential cancer and hazard risks. However, whether employees smoked or not, it was determined that long term exposure to BTEX increased hazard in high-risk areas, such as fuel bays, repair centers and spray painting centers [41]. This was also noted in the current study, where higher concentrations of BTEX were noted in the workshop (where auto-mechanics operate) as compared to the fuel bay (Table 3). This is further illustrated in Table 5 where a greater proportion of auto-mechanics face potential hazard risks, as compared to fuel bay attendants. Colman Lerner *et al.* [32] showed that when compared to many different occupational settings, auto-mechanics and car painting centers showed the highest levels of VOCs, including BTEX.

In both the fuel bay and workshop, there is very little ventilation and no filtration/extraction fans. However, the fuel bay experiences some natural ventilation as air flow occurs through the open doors. This is not the case in the workshop where little to no natural ventilation occurs. This further increases CR and HQ estimates. In Montréal, Canada, BTEX exposure among auto mechanics and painters were within standards; levels were low and did not cause a hazard or cancer risk. However, when both mechanical and natural ventilation systems were used, BTEX concentrations were significantly reduced, as opposed to only natural ventilation system usage [18]. This indicates the urgent need for mechanical ventilation systems to be fixed at the study site and to be maintained properly for such working environments in general.

In addition to lack of ventilation and extraction systems, exposure duration also plays a major role in potential lifetime risks, to both hazard and cancer. Results in this study illustrate that with continuous exposure, CR and HQ exponentially increase, especially for personnel employed for over 30 years (Figure 2). Das *et al.* [8] argued that long term exposure led to increased hazard risk. The researchers found that health related signs were commonly observed in workers employed for more than 5 years at retail petrol pump stations. Workers suffered from neurological symptoms (such as headaches) and eye irritations at these sites. In another study, mean lifetime cancer risks for workers exposed to benzene and ethyl-benzene for 30 years in gas stations in Bangkok, Thailand, was estimated to be $1.75 \times 10^{-4}$ and $9.55 \times 10^{-7}$, respectively [19]. Exposure to these VOCs significantly led to fatigue. These findings are similar to results found in this study (Table 4), where exposure to benzene yielded a mean lifetime cancer risk of $2.19 \times 10^{-4}$.

In addition to smoking habits, ventilation systems and exposure duration, proximity to high levels of BTEX also affects potential risk estimations. Thus, findings revealed that employees placed within offices, further away from direct exhaust emissions, were exposed to slightly lower concentrations of BTEX, and thus experienced lower hazard quotients (Table 5). McKenzie *et al.* [42] determined that the distance from gas wells was significantly associated to the health risks associated with VOC exposure. It was shown that residents < 1km from the gas well were at higher risk of chronic and acute health risks [42]. This was also found by Karakitsios *et al.* [12] in Epirus, Greece, where cancer risk for the general population in close proximity to filling stations increased by 3% to 21%. Thus, many different factors contribute to increased inhalation exposure, and inevitably lead to increased potential health risk.

## 5. Conclusions

The health risk assessment conducted at this site indicates that employees are at risk to carcinogenic effects, and the CR for all employees exceeds the US EPA cancer limits. BTEX concentrations are higher than in other comparative studies. Lack of both mechanical and natural ventilation systems, especially in the workshop, exacerbates the exposure of auto-mechanics and fuel bay attendants. However, despite these findings, confounding factors; such as smoking history, personal medication usage and baseline health status; were not accounted for in this study, and this may skew risk estimations. Future research design should avoid, and/or take into account these confounding factors, as they may affect CR and HR risk estimations.

Overall however, results indicate that ambient concentrations and health risk estimates are generally above international guidelines at the site, and are a matter of concern. This study demonstrates that health risk assessments in conjunction with medical studies (e.g., Keretetse *et al.* [33]) are highly necessary in South Africa and elsewhere, especially in the developing world, to serve as a foundation to amend national exposure limits which will protect employees in high risk jobs.

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## Author Contributions

Raeesa Moolla conducted the experiments, and all authors were involved in data analysis. All authors contributed in the preparation and interpretation of the results. All authors have read and approved submission of the manuscript.

## Conflicts of Interest

The authors declare no conflict of interest.

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# CHAPTER FIVE: SYNTHESIS, GENERAL CONCLUSIONS AND RECOMMENDATIONS

**5.1 Limitations of the study**

**5.2 Synthesis and general conclusions**

**5.3 Future Work**

**5.4 Recommendations**

**5.5 References**

## 5.1 Limitations of the study

Case study research, whilst an appropriate research approach for this study, was not without limitations and problems. A major limitation of this study was a single-site analysis and thus lacks statistical generalisation to sites in different conditions. However, the goal was not one of generalisation, as much to establish whether an occupational risk may exist at diesel refuelling bays, as no such research exists, despite an announcement by WHO in 2012 indicating that diesel emissions are exceedingly harmful to health (World Health Organization, 2012). The results of this study, however, are pertinent to discussions of occupational and risk exposure.

The site, though specific and seemingly worst-case scenario, is not the only such diesel refuelling station in the country. As mentioned in Chapter 1, in South Africa, most bus refuelling stations operate under similar conditions, and experience similar problems (i.e. under cover bays, with little ventilation, lack of filtration systems, little spillage management, use high end 500 ppm Sulphur diesel, etc.). Thus, this research is pertinent as all these stations employ full-time fuel pump attendants and auto mechanics, making this research valuable.

The case study used a single-site because many retail fuelling stations would not grant permission for the study to be conducted. These retail stations stated that they feared service stations would have high concentrations and, thus, they were at risk of being exposed as an occupational hazard. The government-owned bus depot did not have these fears, as research is developmental and constructive, and findings will be used to develop mitigation strategies.

## 5.2 Synthesis and general conclusions

Exposures to BTEX emissions, whether acute or chronic, are potentially harmful to human health. BTEX emissions, as has been noted by researchers, have been linked to numerous adverse health effects, including CNS disorders, respiratory diseases, organ failure, and gastrointestinal disorders (Badjagbo et al., 2010; Capasso et al., 2007; Chen et al., 2011; Demirel et al., 2014; Durmusoglu et al., 2010; Kitwattanavong et al., 2013; Tunsaringkarn et al., 2012; Zhang et al., 2012). Many studies have revealed that human exposures to benzene, even in small concentrations; have increased carcinogenic risk, specifically of leukaemia (Duarte-Davidson et al., 2001; Hein et al., 1989; Huang et al., 2013; Karakitsios et al., 2007a; Moolla et al., 2013; Pilidis et al., 2009; Rasmussen and Khalil, 1983; Rushton et al., 2014; Tondel et al., 1995; Wiwanitkit, 2008). Of major concern is that BTEX emissions worldwide have mainly been investigated in gasoline/petrol fuelling stations, on/near landfills or near chemical industries, but not at diesel stations.

Worldwide, much research has been conducted on diesel particulates and diesel exhaust emissions and their resultant health effects (e.g. Crump, 2014; Kayak and Thompson, 2007; Nightingale et al., 2000; Polosa et al., 2002, 2002; Sydbom et al., 2001; World Health Organization, 2012), however, none of these studies focused on occupational exposure to specifically BTEX concentrations associated with diesel, and health effects thereof. This is of major concern as BTEX compounds are naturally occurring compounds in diesel. The South African government recently stated that diesel, especially high-end diesel (such as 500 ppm Sulphur diesel), is a matter of concern, and PAHs should be monitored (SAPIA, 2008). However, very little on this has been done in the country.

The research results indicate that international occupational exposure limits (OELs) for benzene were exceeded throughout the monitoring period (refer to Chapter 4). Analyses applying Radiello passive samplers (using an aggregate of 14 days exposure) further reiterated this finding, as benzene concentrations were also significantly high at both the refuelling bay and adjacent workshop. This is of concern as full time pump attendants and auto mechanics are employed at the depot.

It is also important to note that BTEX concentrations follow the pattern of refueling activities on site (refer to Chapter 3). Benzene is present throughout the day and night, irrespective of refuelling activities, and/or number of buses refueled daily. However, ethyl-benzene and m- and p-xylene[1] detectable concentrations occur only from 08:00; 30 minutes after refuelling begins at the site, and occurs mainly during refuelling times (i.e. opening times).

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[1] m- and p-xylene co-elute in the GC 955

Diurnal variations of benzene were also noted, with distinct peaks at sunrise and sunset (refer to Chapter 2). This could be attributed to photochemical reactions occurring at these times, increasing benzene concentrations (Rasmussen and Khalil, 1983; Shin and Kwon, 2000). This is noteworthy, as sunrise during winter in Johannesburg is very close to the opening time of the fuel bay, thus attendants are further exposed to high levels of photochemically-induced benzene at this time. Even though the site is under-cover, sunlight still enters the refuelling bay, and thus, photochemical reactions can still occur. Additionally this research further re-iterates findings that $BTEX_{total}$ (where $BTEX_{total}$ is a cumulative amount of individual BTEX species) is significantly higher in the winter season, even in the southern hemisphere, where synoptic climate conditions may differ to atmospheric conditions within the northern hemisphere(refer to Chapter 3).

Furthermore, it was established that concentrations of $BTEX_{total}$ were positively correlated to the volume of diesel dispensed daily (refer to Chapter 2). This conclusion was likewise established in studies analysing the amount of petrol dispensed as compared to BTEX concentrations (e.g. Hein et al., 1989; Karakitsios et al., 2007a, 2007b). It was also noted that on non-work days, $BTEX_{total}$ concentrations were significantly reduced. Ethyl-benzene and o-xylene also indicated a positive correlation with volume of fuel dispensed. It was also observed that there was a negative correlation between $BTEX_{total}$ and temperature. However, patterns in $BTEX_{total}$ cannot be extrapolated for individual compounds as the relative composition of $BTEX_{total}$ changes throughout the day (refer to Chapter 3).

In South Africa, a requirement for vapour recovery systems on refuelling pumps is not legislated for, as is the case in many developed countries (Udonwa et al., 2009). Thus, pump attendants are exposed to vapours emitted during refuelling, as well exhaust emissions, increasing occupational risk. Figure 5.1 further illustrates the close proximity of attendants to exhaust fumes and vapours released from pumps, during refuelling in the bay. Attendants wear protective gloves only during the refueling process (Appendix i), but not while engaged in other activities in the fuel bay. During monitoring, it was noted that diesel spills were not cleaned up correctly (Appendix ii), or adequately, and thus a thin coating of diesel was always present on the floors and workspaces in the fuel bay.

**Figure 5.1: Refuelling of buses by attendants: (a) Exhaust fumes can be seen faintly near the tyre well which adds to the vapours released from refuelling. (b) Close proximity to vapours during refuelling of buses. Protective gloves are worn during refuelling, but removed while engaged in other activities as can be seen in (a).**

Furthermore, due to the poor ventilation and high exposure duration, employees are potentially at risk to adverse health effects (refer to Chapter 4). The average inhalation rate of benzene over the sampling campaign exceeded the US Environmental Protection Agency's chronic inhalation exposure reference concentration. Lifetime cancer risk estimation showed that on average there is a $3.78 \times 10^{-4}$ cancer risk (US EPA recommends that cancer risks above $1 \times 10^{-6}$ are of concern), corresponding to an average chronic daily intake of $1.38 \times 10^{-3}$ mg/kg/day of benzene exposure. Additionally, there were incidences where individuals were at potential hazard risk, from benzene and toluene exposure, which may pose non-carcinogenic adverse health effects for pump attendants and auto-mechanics.

These findings are reiterated in studies conducted in occupational settings where employees were exposed to BTEX concentrations on a daily basis. However, the majority of these studies have been conducted in petrol or gasoline situations (Badjabo et al., 2010; Karakitsios et al., 2007a, b; Kitwattanavong et al., 2013). Nevertheless, concentrations above acceptable criteria for benzene were noted, with cancer risks estimated to be $1.82 \times 10^{-4}$, in a study conducted in Bangkok, Thailand, where exposure of petrol station employees and automobile mechanics to BTEX was evaluated. However, hazard quotients amongst these employees, as is the case at this site, were generally within the acceptable range (Kitwattanavong et al., 2013).

However, as suggested by Badjabo et al. (2010), multiple sources contribute to the occupational exposure of auto mechanics to the BTEX (for example, spray painting, exhaust fumes, cleaning materials), and this should always be considered, as these exposures could increase their potential risk. This is important to consider at this site, as auto-mechanics may be further exposed to these sources (refer to Chapter 4). It should be noted that exposure duration also played a role, as employees employed for 30 years or more showed much greater potential risk, to both cancer and hazard risks. It should be mentioned however, that these calculations are proxy estimates, as they are based on a short monitoring campaign, and thus, the ‘lifetime’ cancer and hazard risks may be over-estimated.

Additionally, the hazard quotient and cancer risks rely heavily on number of years employed. However, it was also noted that employees in areas of increased BTEX concentration levels (i.e. specific workshop areas, refuelling bay, etc.), also potentially face higher risks. Thus, these calculations are proxy estimates, as inhalation exposure will increase exponentially with duration of employment or work locations on site may change (i.e. within the refuelling or workshop bays).

However, due to a lack of comparative national and international occupational exposure limits, identifying exceedances is problematic (refer to Chapter 3). This, however, does highlight the need for the South African government to review guidelines and limits, if proper management and control of air pollution hotspots is to be conducted. Also of significance is the fact that benzene was above international OELs, but within national limits (refer to Chapter 3). The differences noted in international and national OELs of benzene, toluene and xylenes raise many concerning questions. International occupational limits for benzene are significantly lower than South African national standards, while the reverse is true for toluene limits. This in itself is a point of concern as benzene is a carcinogen, even at low level exposure.

Overall, concentrations indicate that occupational exposure at a diesel refuelling bay in Johannesburg, South Africa, placed employees at a significant risk to adverse health effects associated with inhalation exposure of BTEX.

## 5.3 Future work

In South Africa, there is no research related to the amount of aromatic hydrocarbons present in diesel fuel. Thus, analysis of related emissions from exhaust fumes and refuelling activities is based on concentrations measured in these situations, with no guidelines or standards provided by the South African government. In order for scientifically sound research to be conducted, this is imperative. Additionally, the national government need to investigate acceptable levels of BTEX, as this area of research is vital if the health and safety of occupationally exposed workers are to be considered.

A shift needs to also be seen in the perception and importance of occupational health. Organisations and companies, with employees in high risk professions, should allow research to be conducted on site. Personal monitors, testing of blood work and urine samples will further aid health risk analysis immensely, and ensure healthy working environments for workers, enabling higher productivity and lower turn-over rates in the long run. Longer research campaigns, at multiple sites, are also necessary as to enable more accurate predictions, related to health exposure.

## 5.4 Recommendations

Mitigation strategies such as wearing gloves constantly, cleaning up spills in the correct manner and ensuring clean workspaces may decrease exposure risk. In addition, repairing ventilation and introducing air filtration systems will enhance air quality at the sites, and inhalation rates of toxics may be minimised. A major area of concern is the idling of buses during refuelling and repair work. If bus engines are turned off, emission exposure can be greatly reduced in these situations. Thus, a cost-over-health approach should be seen as imperative.

Exposure duration be can further reduced by ensuring that individual offices are properly ventilated, and by allowing meal breaks to be taken outside of the refuelling bay and workshop areas. Any mitigation strategy should ensure that exposure duration (both to fumes from refuelling and from exhausts) is kept to a minimum.

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# APPENDICES

Appendix i: Employees use of gloves during refuelling practices

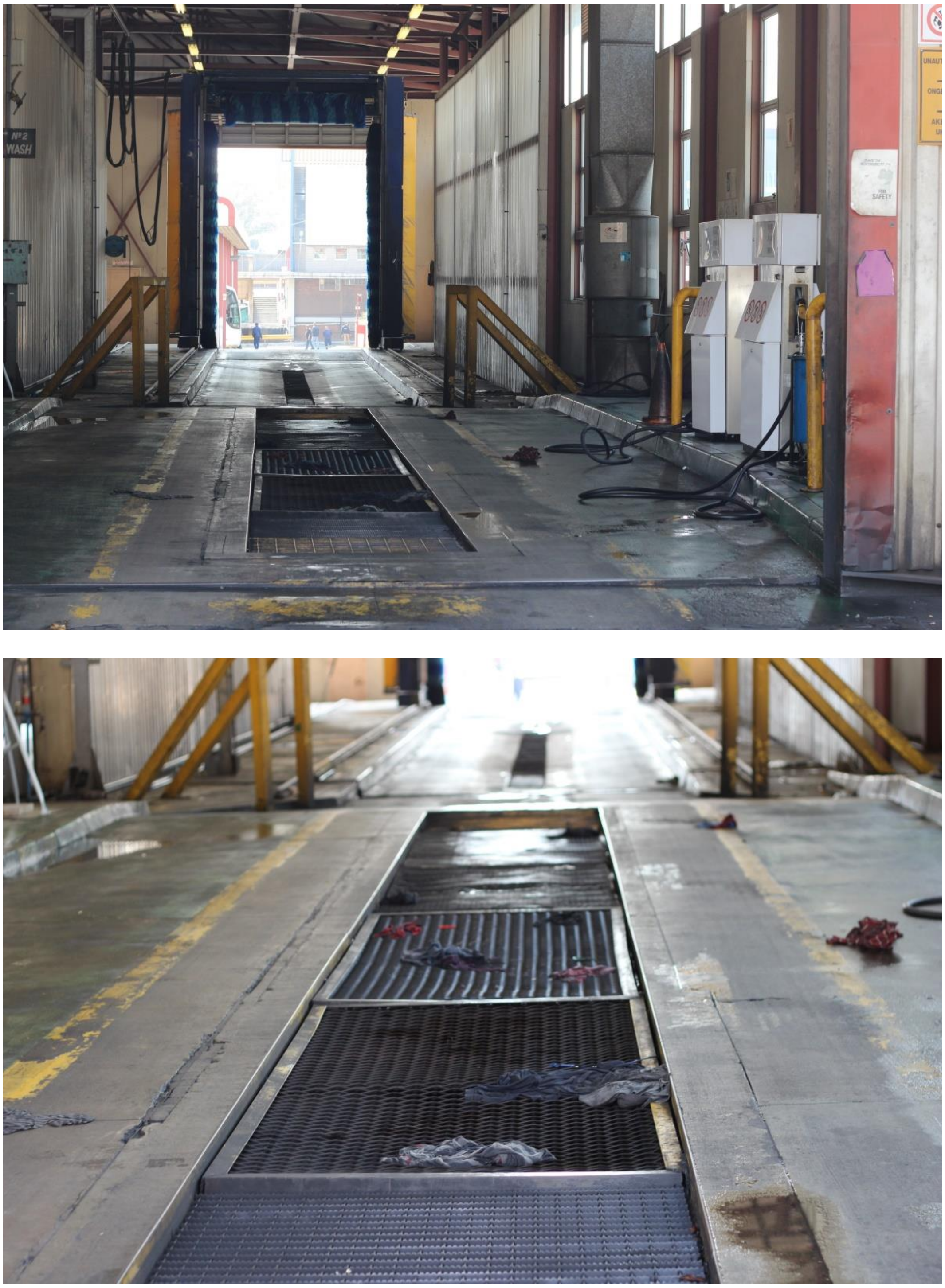
Appendix ii: Spillages of diesel on site in the refuelling bay

Appendix iii: Calculated inhalation rates

# Appendix i

Appendix i: Employees use rubber gloves during refuelling, however, remove them when engaged in other activities, despite refuelling processes still being under way.

# Appendix ii



Appendix ii: Spills on site are not well managed, and are soaked up fibre cloths

# Appendix iii

Table i: Inhalation rates ($m^3$/hour) used to calculate carcinogenic risk and hazard quotient

| Participant ID | Workplace | Position | Inhalation Rate Benzene | Inhalation Rate TEX |
|---|---|---|---|---|
| FBA1 | Fuel Bay | General | 3.71 | 22.97274 |
| FBA2 | Fuel Bay | General | 3.31 | 20.4892 |
| FBA3 | Fuel Bay | General | 1 | 6.20885 |
| FBA4 | Fuel Bay | General | 0.5 | 3.104425 |
| AM1 | Workshop | General | 3.32 | 39.56385 |
| AM2 | Workshop | General | 0.24 | 2.825989 |
| AM3 | Workshop | Office | 5.08 | 27.18813 |
| AM4 | Workshop | Office | 0.12 | 1.412995 |
| AM5 | Workshop | General | 1.13 | 9.489638 |
| AM6 | Workshop | Office | 1 | 7.384181 |
| AM7 | Workshop | General | 0.24 | 2.825989 |
| AM8 | Workshop | General | 0.12 | 1.412995 |
| AM9 | Workshop * | Office | 0.36 | 4.238984 |
| AM10 | Workshop * | Office | 0.24 | 2.825989 |
| AM11 | Workshop | General | 1.19 | 14.12995 |
| AM12 | Workshop | General | 0.59 | 7.064973 |
| AM13 | Workshop | General | 1.9 | 22.60791 |
| AM14 | Workshop | General | 1.3 | 15.54294 |
| AM15 | Workshop | General | 1.9 | 22.60791 |
| AM16 | Workshop * | Office | 0.47 | 5.651978 |

***** Employee based within an enclosed office inside the workshop.