Alternative Techniques to Assess Road Traffic Emissions

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Abstract

Today, one of the most important environmental problems in urban areas is air pollution. According to the World Health Organization, air pollution represents a serious risk to human health in many cities of the world. Road traffic is one of the main sources of pollution in cities. Besides, vehicular emissions are released in close proximity to population, increasing the adverse health effects. Road traffic emissions are highly uncertain in developing countries. Existing techniques to assess this source of pollution are expensive and not always accurate. It results difficult for a city from the developing world to afford these techniques and thus, adequate abatement strategies can not be adopted. It is essential to develop methodologies to reduce these uncertainties to manage air quality more effectively.

This PhD thesis aims to develop and to implement alternative techniques to assess road traffic emissions. This work focuses on the study of Volatile Organic Compounds (VOCs), this family of pollutants have not been sufficiently studied in cities. The developed techniques were tested during an intensive measuring campaign conducted in Ho Chi Minh City (HCMC), Vietnam. The motorcycles are the main mode of transport in HCMC, this means of transport has contributed to the progress of this rapidly developing city. The high levels of air pollution, the elevated number of motorcycles and especially the limited funding available, made HCMC an interesting place to test the developed methodologies.

In a first stage the results of the measuring campaign were used to assess the roadside levels of pollution in HCMC. During the HCMC campaign, 19 C_2-C_6 VOCs were monitored on-line at roadside level together with other important pollutants. Results show that there is a severe air pollution problem in HCMC. The pollutants that are produced by road traffic are identified by means of a Principal Components Analysis (PCA) receptor Model. The PCA shows that all the VOCs monitored (except isoprene) are produced by motorcycles. It is necessary to develop and to encourage the use of alternative modes of transport in HCMC. The use of the on-line roadside pollutants monitoring together with the PCA showed to be a good first approach to assess the road traffic emissions. This is an affordable technique and it is strongly recommended to use it in cities with limited financial means to study the air pollution problem.

This work also presents a new method to estimate road traffic emission factors (EFs). This method is based on a long term tracer experiment conducted during the HCMC campaign. The results of the HCMC tracer experiment were used together with traffic counts and pollutant measurements to calculate the dispersion factors and afterwards the EFs. Estimated EFs for HCMC are within the range of EFs estimated in other studies. Additionally, a Computational Fluid Dynamics Model (CFD) is used to critically evaluate the proposed methodology. The evaluations show that it is possible to accurately estimate the EFs from tracer studies. The methodology proposed here results in an interesting and promising alternative to estimate the EFs.

All the techniques presented in this work offer several advantages. Data obtained serve for different purposes at the same time and their use can provide valuable information for urban air quality assessment. For example, concentrations of pollutants are determined at roadside level, as well as their evolution in time, this information is useful for exposure studies. Roadside monitoring can be used to identify the pollutants that are directly emitted by road traffic. Results from the tracer study can be used to estimate the EFs under real urban conditions and to validate dispersion models which in turn can be used in the future to evaluate abatement strategies for such streets.

Résumé

La dégradation de la qualité de l'air est l'un des problèmes majeurs auxquels les villes d'aujourd'hui doivent faire face. D'après l'Organisation Mondiale de la Santé, la pollution atmosphérique représente un risque élevé pour la santé humaine dans la plupart des villes du monde. Parmi toutes les sources de pollution, le trafic routier contribue très significativement à l'augmentation des risques sur la santé car il est responsable d'une part importante des émissions de polluants qu'il relâche à proximité des populations. Malheureusement, les méthodes permettant de quantifier ce type d'émissions sont à la fois couteuses et pas toujours précises. Dans la plupart des cas, les villes des pays en voie de développement n'ont pas les moyens d'utiliser ces méthodes. Elles sont alors incapables d'évaluer avec exactitude les émissions dues au trafic routier. Ces incertitudes les empêchent d'établir des bilans d'émissions suffisamment fiables et par conséquence de trouver les stratégies les plus efficaces pour améliorer la qualité de leur air. Il est donc essentiel de mettre au point des méthodes permettant de quantifier pour un moindre cout et une précision suffisante les émissions du trafic routier en milieu urbain.

Le but de cette thèse est de développer et utiliser de telles méthodes. Ce travail s'est focalisé sur les Composés Organiques Volatiles (COV) car ces polluants ne sont pas suffisamment étudiés dans les villes. Les méthodes développées dans cette thèse ont été testées lors d'une campagne intensive de mesures s'étant déroulée à Ho Chi Minh City (HCMC) au Vietnam. Les motos y sont actuellement le principal moyen de transport, ces véhicules ont grandement contribuées au développement de la ville durant les deux dernières dizaines d'années. Les hauts niveaux de pollution atmosphérique, le nombre élevé de motos et les faibles moyens financiers disponibles font de HCMC un emplacement particulièrement approprié pour ce travail de recherche.

Dans une première étape, une campagne de mesure a permit d'évaluer les niveaux de pollution à HCMC. Durant cette campagne, 19 COV (C_2-C_6) ainsi que les polluants NO et $PM_{2.5}$ ont été mesurés en continu dans une rue. Les résultats montrent que HCMC est confrontée à un grave problème de contamination par les polluants atmosphériques. L'origine de ces polluants a été identifiée à l'aide d'une méthode d'Analyse en Composantes Principales (ACP). L'ACP montre que les COV

(à l'exception de l'isoprène) sont produits par les motos. Ceci permet de conclure qu'il est nécessaire d'encourager l'utilisation d'autres types de transport moins polluants à HCMC. La mesure en continue de polluants atmosphériques sur des sites urbains associée à l'utilisation d'ACP apparait être une excellente première approche pour évaluer l'origine des émissions dues au trafic. Cette méthode étant très accessible, son utilisation est fortement recommandée dans des villes ne pouvant dédier que de faibles moyens financiers à l'étude de la qualité de l'air.

Ce travail présente également une nouvelle méthode pour les facteurs d'émissions (FE) (i.e. quantité de polluants émise par les véhicules par km parcouru). Cette méthode est basée sur l'émission en continue d'un traceur passif, de comptages de véhicules et de mesures de polluants. Elle permet de calculer les facteurs de dispersion des polluants puis les FE. Il apparait que les FE ainsi estimés pour HCMC sont du même ordre de grandeur que ceux estimés dans d'autres études. De plus, un modèle de Mécanique des Fluides Numérique a été utilisé pour évaluer la qualité de la méthode proposée. Les simulations ont montrés qu'il est possible d'estimer avec une bonne précision les FE grâce à l'émission du traceur passif. La méthodologie développée dans cette thèse se montre donc comme une alternative à la fois intéressante et prometteuse pour trouver les FE.

Toutes les méthodes présentées dans ce travail offrent plusieurs avantages. Ces méthodes peuvent donner plusieurs types de réponses en même temps et fournissent ainsi des informations utiles pour la gestion de la qualité de l'air. En effet, les mesures effectuées au niveau de la rue permettent de déterminer l'évolution temporelle des concentrations de polluants. Cette information est utile pour étudier l'exposition de la population à la pollution atmosphérique. Elle peut également être utilisée pour identifier les polluants directement émis par le trafic routier. Enfin, l'émission d'un traceur passif permet d'estimer les FE en situation réelle et de valider les modèles de dispersion qui peuvent servir à évaluer l'efficacité des stratégies de réductions de la pollution atmosphérique.

Mots clés: Gestion de la qualité de l'air; émissions en situation réelle; incertitudes des inventaires d'émissions; chromatographie en phase gazeuse; Composés Organiques Volatiles (COV); modélisation de la qualité de l'air; Mécanique des Fluides Numérique; validation de modèle atmosphérique de dispersion.

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CHAPTER 1

INTRODUCTION

The population living in urban environments has rapidly increased in the last decades. In 1950 there were 83 cities with more than one million inhabitants. Today there are more than 350. According to the United Nations, near 50% of the world's population live in urban areas, today, this number will increase to about 70% in 2050. Urban population in developing countries will double in the next 20 years.

People living in urban areas require a large amount of resources whose use produce wastes with environmental consequences from urban to global scale. Today one of the most important environmental problems in urban environments is air pollution. According to the World Health Organization, air pollution represents a serious risk to human health in many cities of the world. Thus, the protection of the human health should be the main objective of the urban air quality management. Most of the exposed people live in cities of developing countries. Malnutrition problems and the limited access to health services make the dwellers of such cities more vulnerable to air pollution.

The basis of an urban air quality management program is the correct identification of sources of pollution. Different techniques exist to identify the sources but these tools are cost expensive and not always accurate. It results difficult for a developing country to afford the implementation of such tools and thus adequate abatement strategies can not be adopted. It is essential to develop cost effective methodologies to manage air quality more efficiently in such cities.

Road traffic is one of the main sources of pollution in cities. Besides, vehicular emissions are released in close proximity to population, increasing the adverse health effects. In developed countries, modern vehicular combustion and emission control technologies are used; new and efficient transport systems exist and thus the air quality has improved significantly in the last years. In developing countries old

vehicle technologies are still used and massive transport systems are inadequate or inexistent and therefore most of these cities present high levels of air pollution.

The aim of this PhD thesis is to develop and to implement alternative and cost effective techniques to assess the road traffic emissions. The developed techniques were tested in Ho Chi Minh City (HCMC, Vietnam). Vietnam is a developing country with one of the fastest developing economies of the world. HCMC is one of the largest cities of the Southeast Asia. In the last yeas the city has developed rapidly and this development increased the demand for road transport. Due to the lack of an adequate massive transportation system, the motorcycles became the main mode of transport in the city and also one of the main sources of pollution. The economical constrains and especially the large number of motorcycles difficult the use of traditional techniques to study the air pollution. Therefore it is an interesting place to test the developed methods.

Most of the air quality research developed in many cities of the world has been focused on the study of particles and ozone. But the number of researches dedicated to the study of other important air pollutants like Volatile Organic Compounds (VOCs) is limited. VOCs are an important family of pollutants not only because they play a key role in atmospheric chemistry, but also because some VOCs have toxic health effects. Part of the research of this thesis is devoted to the study of VOCs in HCMC. The work is based on an intensive measuring campaign that took place in the centre of HCMC from January to March 2007. A state-of-the art on-line VOCs gas chromatograph together with other cutting edge devices were used for the air pollutants measurements.

This work is organized as follows:

Chapter 2: Presents and outlines the actual state-of-the-art in the field of road traffic and urban air quality. First, Key topics related to the urban air quality are shown. After, the air quality management and the assessment tools are presented and evaluated. The importance of road traffic in the urban air quality is discussed and the current state of research concerning the available techniques to estimate road traffic emission factors is assessed.

Chapter 3: In this chapter the results of the HCMC measuring campaign are presented. 19 VOCs, NO and $PM_{2.5}$ were measured on-line at roadside level during the campaign. The pollutants that are produced by road traffic and especially by motorcycles are identified by means of the Principal Components Analysis receptor model (PCA). The information collected is also used for other important urban air quality assessments. The VOCs ozone formation potential is calculated and pollutants concentrations are compared with the results from other studies.

Chapter 4: The design and test of an innovative technique to estimate road traffic emission factors (EFs) is presented in this chapter. EFs are estimated from a long term tracer experiment and from roadside measurements. EFs for 15 VOCs and NO are reported and evaluated. The estimated EFs are compared with the EFs reported in other studies.

Chapter 5: In this chapter the results of the tracer experiment are used to evaluate the performance of a Computational Fluid Dynamics (CFD) model. The method presented in the previous chapter is also critically evaluated by means of the CFD model. Different alternatives to improve the method are proposed and assessed with the model.

Chapter 6: Present the conclusions from this PhD thesis and give recommendations for further research works.

CHAPTER 2

ROAD TRAFFIC AND
AIR QUALITY IN CITIES

Motor vehicles are an important part of the modern life. They allow the easy contact between people and open the horizons for personal and professional activities. Road transport has also greatly contributed to the development of the world. The economics of entire regions depends on the easy access to people and goods provided by road transport. Unfortunately, these positive aspects are accompanied with the detriment of the environment produced by the hazards released by the vehicles.

In this chapter the state-of-the-art in the field of road traffic emissions and air quality is discussed; the scientific research needed is also presented. First, key topics related to the actual urban air pollution problems are shown. After, the air quality management and the assessment tools are presented and evaluated. Subsequently, the available techniques to estimate road traffic emissions are reviewed.

2.1. URBAN AIR QUALITY

Nearly half of the world's population in 2000 lived in urban areas, and the number of urban dwellers is expected to grow by 2% per year during the coming three decades (Molina and Molina, 2004). These concentrations of people and their activities have consequences at urban, regional, continental, and global scales. One of the main consequences of urbanization is the deterioration of the urban air quality.
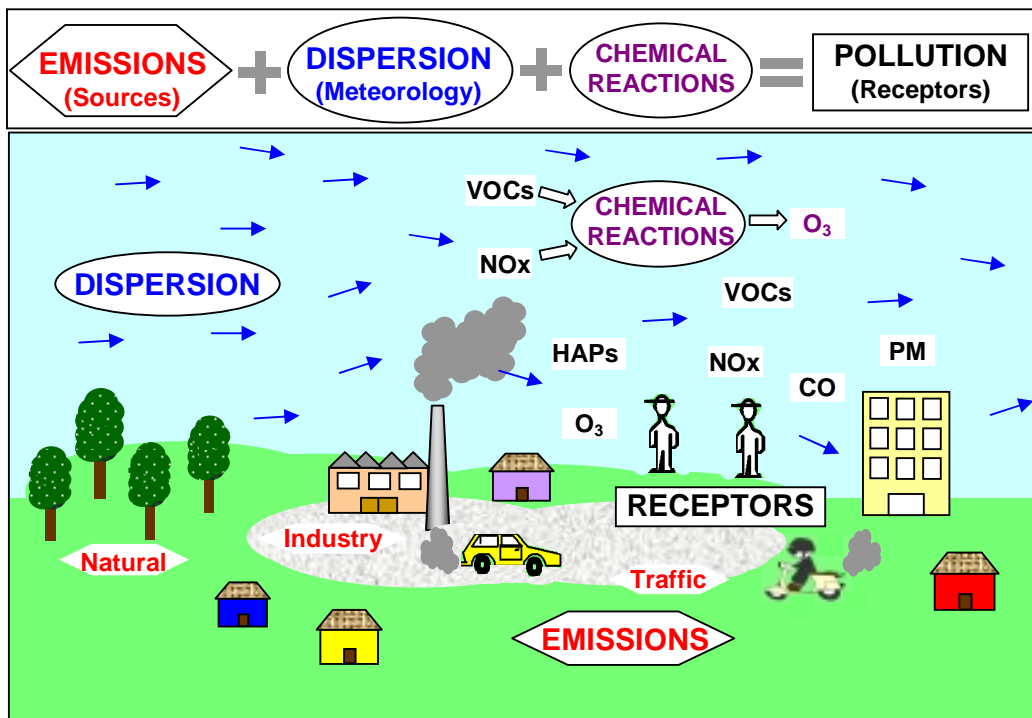
Figure 2.1. Main factors involved in the urban air pollution.

Figure 2.1 presents the 3 main factors involved in the urban air pollution problem: the emissions, the dispersion and the chemical transformations. Air pollution levels presented in cities depend on the different possible combinations of these factors.

Air pollution in cities comes from many different emission sources (USA-EPA., 2009): stationary sources such as factories, power plants, smelters; and smaller sources such as service stations; mobile sources such as cars, buses, planes, trucks, and trains; and naturally occurring sources such as volcanic eruptions, windblown dust and woods, all contribute to air pollution.

The different pollution sources in a city emit a large number of pollutants. More than 3 000 different anthropogenic pollutants have been identified, most of them organic. Combustion sources, especially motor vehicles, emit about 500 pollutants. However, only the impacts of about 200 pollutants have been investigated; the ambient concentrations have been measured for an even smaller number (Fenger, J., 1999).

Air pollutants can be classified in two groups:

- ***Major air pollutants***: comprising sulphur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), lead and ozone (O_3), particulate mater (PM)

- Hazardous air pollutants (HAPs): Comprising chemical, physical and biological agents from different types and that are known or suspected to produce serious human health effects

The HAPs are present in the atmosphere in much smaller concentrations than the major air pollutants, but they are, due to their high specific activity, nevertheless toxic or hazardous. Both in scientific investigations and in abatement strategies HAPs are difficult to manage not only because of their low concentrations, but also because they are in many cases not studied. HAPs are subdivided into various groups such as, particulate mater (PM), dioxins and metals. Volatile Organic Compounds (VOCs) are also considered as HAPs.

The pollutant dispersion plays an important role in the urban air quality, i.e. two cities having the same levels of emissions may have completely different pollution problems. The dispersion is directly related to the meteorological conditions present in the city. The main meteorological variables involved are the wind speed, wind direction, precipitation, ambient temperature and mixing layer high. These meteorological variables are highly associated to the topography of the region in which the city is located.

The chemical atmospheric reactions are also an important source of pollution. Once in the atmosphere, the pollutants chemically react with other compounds to produce other pollutants. The pollutants that are directly emitted from the sources are known as **primary pollutants**. The pollutants that are chemically formed in the atmosphere are known as **secondary pollutants**. There are several secondary pollutants, some of the most important in urban environments are particles (PM) and ozone.

High levels of pollutants in cities cause harm to people or other living organisms, or damage the environment. The impact to the human health is the main concern in the regulation of urban air quality. Several studies have linked air pollution to increases in human mortality and morbidity rates. The World Health Organization states that air pollution is a major environmental risk to health and is estimated to cause approximately 2 million premature deaths worldwide per year (WHO, 2009). More than half of the burden from air pollution on human health is borne by people in developing countries.

A large number of studies have been devoted to find the links between major air pollutants like ozone and PM to human health. But the studies developed to assess the levels of HAPs and to link these pollutants to human health are limited. Available studies show significant associations between some HAPs and adverse health consequences like cancer, neurological, respiratory, reproductive, and developmental effects. However, the human health effects from a large number of compounds remain still unknown.

2.2. AIR QUALITY MANAGEMENT

Air pollution is a complex problem. It involves a large number of variables, there are typically thousands of sources in a city and the pollutants once emitted are subject to numerous physical and chemical processes. In addition, the relationships between the resulting pollutants concentration and all the variables involved are by no means linear.

Different socio-economical aspects are involved in this problem which means that the air pollution is site specific. Then, the solutions that may be adopted in a city may not be useful in another city with other socio-economical conditions.

The design of effective abatement strategies requires a good understanding of the problem. Scientists together with environmental authorities have developed a set of tools that help to understand the origins and fates of the pollution in cities. These tools have been successfully used to assess pollution and to propose efficient abatement strategies but due to the complexity of the problem there are still concerns about the pollution levels even in many cities of developed countries.

Figure 2.2 presents the available tools to manage air quality. These tools include: air quality monitoring, emission inventories, air quality modeling and receptor modeling. Although these techniques can be used separately to adopt abatement strategies, the most convenient way to manage air quality more efficiently is to use all of them together. However, it is not always possible due to economical and technical constraints.
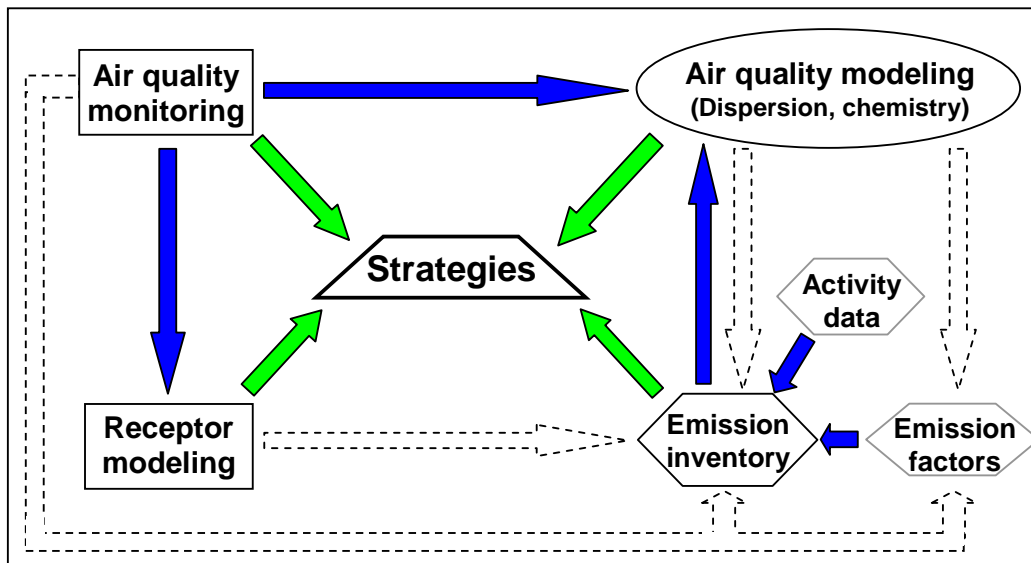
Figure 2.2. Available tools to manage air quality in cities. Blue arrows: Information used as input for other tools; Green arrows: Information used to adopt or to evaluate the strategies; Doted arrows: Tools used to evaluate or to improve other tools.

Available techniques have been also designed to produce input information for other tools. In the last years innovative methods have been designed to evaluate or to improve existing techniques. For example, air quality models have been recently used to evaluate the accuracy of the existing emission inventories (Zarate et al., 2007)

The current state-of-the-art together with a critical assessment of these tools is presented below.

2.2.1. Air quality monitoring

Air quality monitoring is considered the first step in an air quality management program. It has been conducted to measure the urban or the rural background, and the roadside levels of pollution. Urban and rural background monitoring has been developed to establish the ambient levels of pollution, to estimate the level of pollution exposure, to evaluate the effectiveness of abatement strategies and to identify the sources of pollution. Roadside monitoring has been developed to evaluate pollution exposure, to study their diurnal and seasonal variations and to link these variations to the pollutants produced by road traffic.

Different techniques can be used to monitor the air quality; they can be classified as manual and automatic. Manual methods consist in collecting air quality samples at the selected site. These samples are stored in previously adequate containers (i.e. tube or a canister), and later transported to the laboratory for analysis. Manual techniques are also used to measure the levels of certain pollutants which can not be measured automatically like PM species. This method is a suitable technique to assess in an easy and affordable way the air pollution levels during a short period of time. However, in the long term, the use of these technique results inefficient.

Automatic monitoring devices allow the real time in situ determination of air pollutants levels. Automatic techniques allow the acquisition of a large amount of data. This information is useful to determine the daily and seasonal variations of pollution and to relate the pollutants to their sources. In contrast, these techniques are expensive usually require trained personnel to properly operate the devices (Molina and Molina, 2004).

Air pollution levels have been routinely monitored in developed countries during several years (Demerjian, 2000). More than 4 000 urban monitoring stations exist in North America and several hundreds of stations also exist in Europe and in other developed countries. The information collected has been crucial to improve their urban air quality. In the last 10 years devices to measure the levels of selected HAPs in real time have been installed in such countries. For example, important pollutants like ozone precursors started to be automatically measured in the monitoring networks. However, only few studies have been conducted to analyze the long term databases of HAPs collected in developed countries (McCarthy et al., 2007).

The situation in developing countries is different. Air quality monitoring in the cities of such countries is limited and long term records are rare. Typically only major pollutants are routinely monitored in larger cities whereas HAPs levels are unknown in most of the cities of the developing world. In some cities the local environmental authorities have purchased the necessary equipment, and even different international cooperation agencies have donated modern automatic equipment. Nevertheless, these devices are usually expensive to maintain and they require highly qualified technical support which is usually not available in such countries and thus these devices are not used.

2.2.2. Emission inventory

An emission inventory is an accounting of the amount of pollutants that are released from the different sources into the atmosphere of a region during a given time period. The development of a complete emission inventory is also an important part of an air quality management program.

Emission inventories are used to identify the main sources of pollution, to state environmental priorities, and to assess the potential benefits of different strategies and plans. Emission inventories group the sources in different categories, i.e. mobile sources (road traffic), stationary sources and area sources. The major pollutants and in some cases HAPs are included in the emission inventories.

The basic model for the quantification of the emissions is the product of two variables:

$$E = EF * A \qquad\qquad (2.1)$$

Where E is the total emission, EF is the emission factor and A is the activity data. An EFs is a representative value that relates the quantity of a pollutant released to the air with the activity associated with the release of that pollutant (EEA, 2007). EFs are expressed as the weight of pollutant divided by a unit of weight, volume, distance, or duration of the activity emitting the pollutant. Such factors facilitate the estimation of emissions from various sources of air pollution.

The European and the US Environmental Protection Agencies have developed local emission inventories for about 30 years. Although several million dollars have been invested, and despite all the efforts made to improve the emission estimates, these emission inventories are still subject to uncertainties. Parrish (2006) carried out a critical evaluation of the US on-road vehicle emission inventories. He used ambient pollutants measurements and fuel based emission estimations for this analysis. This evaluation showed that the emission estimates are not clearly converging to more accurate and certain estimates. Results also showed that the CO vehicle emissions are overestimated by a factor of 2.

Parrish also used benzene and acetaldehyde ratios to evaluate the inventory speciation of VOC. Figure 2.3 shows a comparison between the inventory and the observed ratios of these two species during a period of 25 years in the US. Although recent studies show that these two species are mainly emitted by road traffic, a large difference between the total and the on-road emission ratios is observed. Moreover, the inventory ratios for total emissions are a factor of 3-4 higher than the ambient values. On-road emission ratios are also higher than those from the observations. Further the temporal trends are not in agreement. This study concluded that there is a critical need for a reevaluation of the VOC speciation emissions throughout the US.
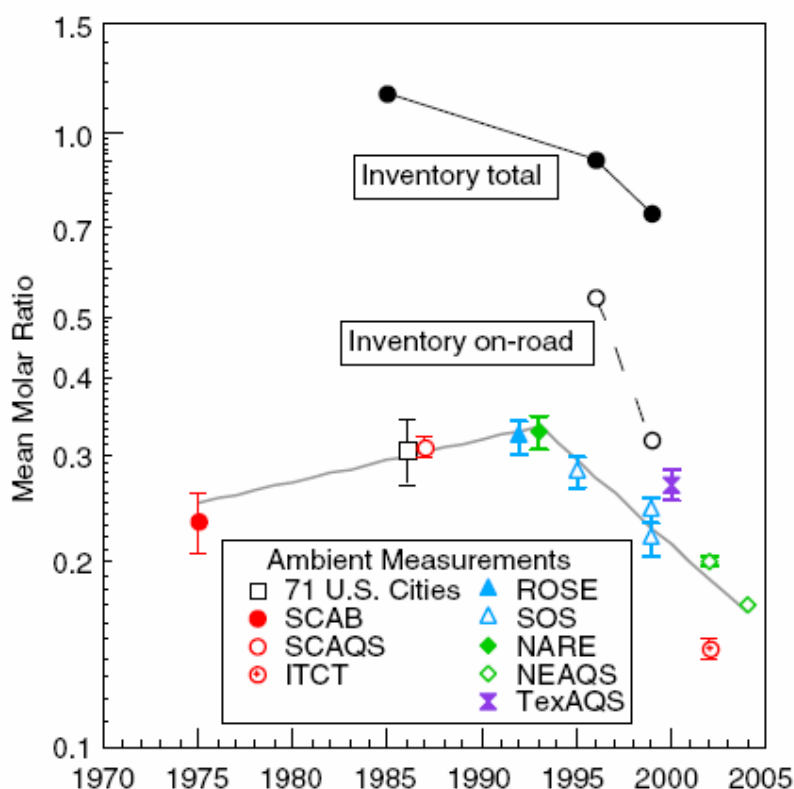
Figure 2.3. Temporal trends (25 years) of observed ambient benzene to acetylene ratio from field study data compared to inventory ratios (semi-log plot). California: circles; Southeast US: triangles; Northeast US: diamonds; Texas: TexAQS. The solid grey line is the trend of the measurements ratios (Parrish, 2006)

Many cities from the developing countries do not count on information about their emissions. In the best cases, the policies to improve air quality are based on incomplete emission inventories. There, only major pollutants are covered but the amount of the emission of other relevant pollutants like PM or VOC species and their

trends are unknown. In these cities, due to technical and economical constraints, EFs from developed countries are adopted although they have been estimated in completely different conditions. Moreover, information about activity data is usually not available or is not well organized; adding even more uncertainties to the emissions inventories.

2.2.3. Air quality modeling

Dispersion models use mathematical expressions to represent the dispersion and some times the chemical transformation of pollutants in urban environments. These models estimate the resulting pollutants concentrations under given emissions and meteorological conditions. There are different dispersion models with different degrees of complexity and for different scales ranging from entire urban areas or regions to small urban microenvironments.

Medium scale models or mesoscale models (i.e. CALPUFF, ADMS, TAPOM, CHIMERE, CMAQ) are used to simulate the physical and, in some cases, the chemical processes that occur in an urban area. These models require input information like the meteorological conditions present in the place and the emission inventory. Mesoscale models are mainly used to evaluate the effectiveness of strategies to reduce the pollution in a city before they are implemented. These models have been also used to evaluate the accuracy of the existing emission inventories (Zarate et al., 2007).

Small scale models, or microscale models, are used to simulate the processes involved in the dispersion and transformation of pollutants over part of a city. Several models have been developed to simulate the pollutants dispersion and transformation in street canyons (urban streets flanked by buildings on both sides, Vardoulakis et al., 2003). They are of interest because high levels of pollution have been detected inside these urban environments.

Street dispersion models calculate pollutant concentrations in two different ways. A set of models called *operational models* solve analytically a set of parametric semi empirical simplified equations. Computational Fluid Dynamics models (CFD) solve numerically a set of differential equations that describe in detail wind flow and pollutant dispersion.

Operational models (i.e. STREET-SRI, CALINE4, OSPM, AEROLIUS, etc), are specially designed to produce time series of pollutants concentrations near urban streets (Vardoulakis et al., 2003). The main advantage of these models is that they are able to produce results with a reduced number of information and with minimal computational requirements. The main weakness of these models is that they are based on a number of empirical assumptions and parameters that might not be applicable to all urban environments. For this reason, they should be recalibrated against field measurements if they are to be applied to new locations.

CFD modeling is a powerful technique spanning a wide range of industrial and more recently environmental applications. The main advantage of these models is that they can reproduce the entire flow and concentration fields for any street configuration (i.e. FLUENT, MISKAM, ENVIMET). The main limitation of CFD models is the large amount of data required. The long computational time needed for the simulations is another important limitation. There is also a lack of validation of these models against physical modeling results or field measurement data (Li et al., 2006). In addition, these models have been typically validated using wind tunnel measurement data but the number of validation studies using measurements under real urban conditions is limited.

2.2.4. Receptor modeling

Receptor models (CMB, PCA, UNIMIX, PMF, etc.), also known as source apportionment, use mathematical or statistical procedures to identify the source that contribute to pollution in a receptor point. Dispersion models use the best available emissions and meteorological data to predict the pollutant concentrations. Receptor models do not need any emission or meteorological data. Instead, they use the abundance of chemical components measured at receptors and sources to quantify the source contributions. Anyhow, both modeling techniques have strengths and weaknesses that compensate each other and then both techniques should be used in an urban air quality management program.

The main advantage of receptor models is that they can show the importance of emission sources directly without the need of emissions and meteorological data or chemical transformations mechanism. Moreover, a receptor modeling study produces additional information that can be used for other air quality assessments like air quality standards compliance or epidemiology studies. Receptor modeling studies

have been also used to cross check the reliability of existing emission inventories. This modeling technique has found large discrepancies between ambient measurements and emission inventories for vehicle exhaust and road dust (Watson et al., 2002).

One of the main weaknesses of receptor models is that they assume the pollutant species are not reactive or react slowly. This assumption generates uncertainties on the source identification especially for pollutants that are reactive like some VOC species. Another limitation of receptor models is that they need a relatively long dataset to better identify the sources. The sample number must greatly exceed the number of selected species to obtain robust results. It is recommended to collect at least 50 samples (Guo et al., 2004). It results difficult and cost expensive to collect this number of samples especially for pollutants that can only be monitored manually like PM species.

2.3 ROAD TRAFFIC EMISSIONS

2.3.1. Importance of road traffic in the urban air quality

In 2007 there were about 810 million motor vehicles in the world and about 100 million motorcycles (Autonews., 2008). These vehicles use about 260 billion gallons of gasoline and diesel fuel yearly; most of these fuels are burn in the urban environments.

Despite the use of advanced combustion and emission control technologies, road traffic is still an important source of pollution in cities of developed countries, mostly due to their large number of vehicles. Vehicle fleet in developing countries is typically older, usually emission control devices are not used and they are not required for the environmental authorities. Therefore, road traffic is also one of the main sources of pollution in such countries.

The main difference between road transport and other sources is that road transport sources release their emissions in a very close proximity to human receptors. This reduces the opportunity for the atmosphere to dilute the emissions which would render them less likely to damage human health. Furthermore, in most cities,

concentrations of vehicle exhaust are significantly enhanced by the fact that many roads have buildings alongside (Colvile et al., 2001). The effect of such buildings is to shelter the road, reducing the wind speed at the source of emissions by as much as an order of magnitude relative to that on an open road

Despite road traffic is an important source to consider in the urban air pollution problem, EFs and activity levels for this source category are highly uncertain (Molina and Molina, 2004) It is essential to improve existing methodologies or to develop new methods to accurately estimate road traffic EFs.

The previous paragraph makes evident the great importance to accurately estimate the vehicle emission factors (EFs) in a city. However, they are difficult to estimate because there are several factors involved.

2.3.2. Variables affecting the vehicle emissions

The main variables affecting the emissions produced by a single vehicle are:

- **Vehicle type:** Vehicle technology, size, age, mileage, and fuel type used.
- **Vehicle operation:** Travel speed, acceleration, deceleration, vehicle load and the maintenance record.
- **Local conditions:** Ambient environmental temperature, altitude above the sea level and road slope.

Moreover, the pollutants are not only emitted from the exhaust of the vehicle (exhaust emissions). There are also non-exhaust emissions; VOCs from the fuel evaporation, dust resuspension and particles from tires and brakes wear are also emitted by a car. Non exhaust emissions are an important source to consider (Mellios et al., 2006).

Additionally, since it is not possible to measure the emissions to the entire vehicle fleet of a city, a representative sample must be taken. There is not a standard methodology to determine the size of this sample. Instead, the number of vehicles selected for the measurements depends on the available budget.

2.3.3 Available techniques to estimate road traffic emission factors

The suitable methodology to correctly estimate the vehicle EFs should be able to consider all the variables mentioned above, for a representative sample of the fleet analyzed, and for a large number of pollutants.

Two different approaches have been developed to estimate the vehicle EFs in a city. These approaches are known as bottom-up and top-down. Bottom-up is considered as the traditional method whose most representative techniques are chassis dynamometer and on board emissions measurements. The most used top-down techniques (also known as real world emission monitoring) are the tunnel studies and the inverse modeling.

A brief description of these techniques together with their main strengths and weaknesses is presented below.

Bottom-up methods

- Chassis dynamometer. In this method a dynamometer is used to mimic the urban driving conditions in a laboratory (figure 2.4). During the emission measurements, a vehicle is driven in a rolling road. The dynamometer provides simulated road loadings to the vehicle, at the same time the exhaust emissions (some times the non-exhaust emissions) are monitored. The loadings are obtained from a predefined driving cycle. The driving cycle is supposed to represent the way a vehicle is driven in a typical urban trip. Different driving cycles have been developed for different cities of the world.
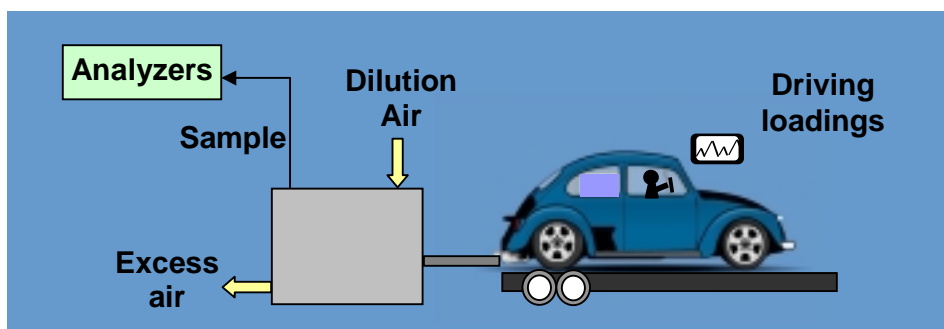

Figure 2.4. Estimation of emission factors in a chassis dynamometer.

Strengths: By using this method the emission measurements are obtained under standardized laboratory conditions. Dynamometer methods are useful techniques for comparative testing. Since the measurements are carried out under controlled conditions, it is possible to evaluate the impact of emission reduction strategies (i.e. effects of different fuels, additives, engine technologies and emission reduction devices). Dynamometer test are also a useful technique to define environmental regulations.

Weaknesses: One of the main limitations of this technique is that most of the driving cycles don't represent the real urban driving behavior. In addition, all the equipment required for the measurements are cost expensive. Another important weakness of this method is the limited number of vehicles that can be tested. Ropkins et al. (2009) states that only 2 to 20 vehicles can be measured in a chassis dynamometer per day.

- On board emissions. In this method the emission monitoring devices are installed in the trunk or in the seats of the vehicle. The vehicle is driven along a predefined circuit that represents the typical local urban driving conditions. During the circuit, a sample of the exhaust gases is continuously analyzed on real time or almost real time to determine the pollutants concentrations. To estimate the emission factors it is necessary to measure the exhaust air flow. This flow is either measured at the pipe or calculated indirectly using engine parameters or inflow fuel and air.

Strengths: This technique presents most of the strengths the dynamometer technique offers but at a lower cost. Moreover, the EFs are estimated at real urban conditions.

Weakness: The EFs are derived from a smaller number of vehicles. This is because in addition to the time needed for the emission test, additional time is spent mounting, setting up and dismounting the devices. Even an experienced and well organized team can only measure as much as 10 vehicles per day with an on board device. Other limitation of this method is the reduced number of pollutants that can be assessed. Typically, only the EFs for the major pollutants can be estimated. Additionally, it can only estimate the exhaust EFs. As it has been explained before, the non exhaust EFs are an important parameter to consider.

Top down techniques

- Tunnel studies. In this method, air quality monitoring devices are deployed inside the tunnel and in its surroundings. At the same time the vehicles is also continuously registered. The average EFs are derived from a simple mass balance. It is also possible to estimate the EFs for individual vehicle categories by using regression analysis (Cheng et al., 2006).

Strengths: Roadway tunnel studies have been the most used top down technique to estimate vehicle EFs because they can be derived in a practical way and at a low cost. Moreover, the estimated EFs are estimated from a large number of vehicles (4 000 – 25 000 veh/day, Ropkins et al., 2009).This technique also consider the exhaust and non exhaust emission. In addition, the EFs for a long list of pollutants including several HAPs can be estimated.

Weakness: The EFs are estimated under site specific conditions. They only represent the vehicle fleet, the speed, and the road slope present in the tunnel. Besides, it is not always possible to find a vehicular tunnel in a city. Usually these tunnels are located outside the cities. There, the driving behaviors are different to the typical urban driving patterns. Moreover, despite it is possible to derive the EFs for different vehicle categories, these EFs are still an average of the category. In addition, by using this method, and since the EFs are measured in situ under real conditions, it is not possible to evaluate emission reduction strategies or technologies as it is possible in bottom up techniques.

- Inverse modeling. A linear relation between the EFs (q), the pollutants dispersion (F_i), the number of vehicles (N_i), the roadside (C_i) and the background concentration (C_b) is used to estimate the EFs (figure 2.5). Pollutant concentrations are measured at roadside and sometimes also at the background level. The pollutants dispersion is computed by using either a microscale operational or either a numerical CFD model. The linear regression of the C_i vs. F_i*N_i plot from all data available gives from the slope the traffic emissions factor and from the intercept the average C_b (Palmgren et al., 1999).
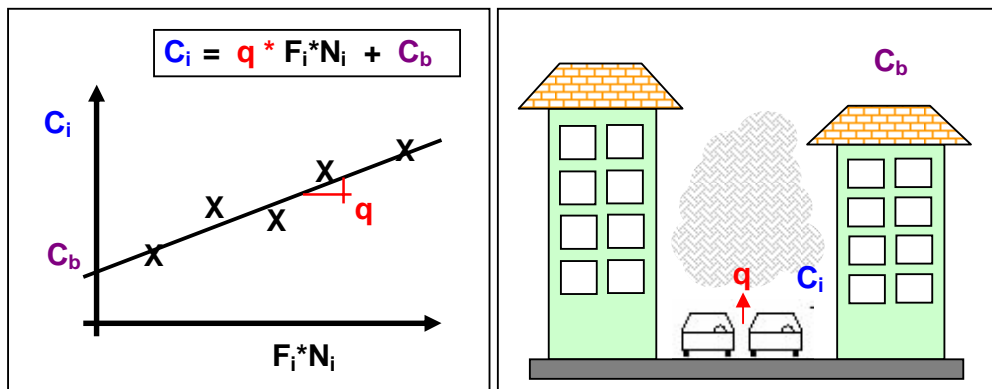
Figure 2.5. Estimation of the emission factors from the inverse modeling

Strengths: The most significant advantage of this method is that the EFs are also estimated under real urban conditions from a representative number of vehicles (4 000 – 25 000 veh/day, Ropkins et al., 2009). The EFs for a large number of pollutants can be also obtained. This technique also considers the exhaust and the non exhaust emissions in the estimations.

Weakness: Here the accuracy of the calculations depends on the ability of the model to simulate the dispersion of the pollutants. Intercomparison studies have shown large discrepancies between microscale operational models fed with the same inputs parameters (Vardoulakis et al., 2003). Moreover and despite all the efforts made in the last years, at the moment CFD models have not been sduficiently validated using information obtained from real urban conditions (Li et al., 2006).

2.4. CONCLUSIONS

In this chapter, the state-of-the-art in the field of road traffic and urban air quality was reviewed and discussed. The basis of a successful air quality management program is the correct identification of the sources of pollution. Road traffic is one of the main sources of pollution in cities but despite all the efforts made in the last years by the scientific community, there are still several scientific gaps and uncertainties associated to this source, which limit the correct understanding of the problem.

The available air quality management tools were critically assessed. In developing countries only few atmospheric pollutants concentrations are monitored despite

hundreds of pollutants are released. In such countries, the levels of important hazardous air pollutants are unknown.

Not all the sources of the pollutants present in the atmosphere have been identified. Receptor models appear as an interesting alternative to identify the sources. They can show the importance of the emission sources directly without the need of additional information like emissions inventories, meteorological data or chemical transformations mechanisms. Several receptor modeling studies have been carried out to identify the sources of particles but very few to identify the sources of other important hazardous pollutants (i.e. VOCs). This research is needed not only because of the important impacts that these HAPs have on the environment but also because it provide useful information for assessing urban air quality.

The emission inventory has been one of the most used tools to identify the sources of pollution but there are still several uncertainties associated to road traffic emission factors (EFs). Two approaches exist to estimate the emission factors which are known as bottom up and top down. Both approaches offer several advantages and face weaknesses that limit the correct estimation of the EFs.

One of the main weaknesses of bottom up methods is that the EFs are estimated for a limited number of pollutants and from a small vehicle sample. One of the main limitations of the top down techniques is that the EFs are estimated under site specific conditions. Recently, an alternative top down technique called inverse modeling was developed to tackle this limitation. However, the accuracy of this technique depends on the ability of the models to reproduce the dispersion of the pollutants. Most of the available dispersion models are not applicable to all the urban environments. Advanced CFD models are designed to be applicable in any urban configuration but at the present they have not been sufficiently validated using information obtained from real urban conditions.

At the moment, there is not a unique technique able to accurately estimate the traffic EFs. It is recommended to use both, bottom up and top down techniques to estimate the EFs. However it is not always possible especially in developing countries were the funding are limited. It is essential to develop and to test suitable methodologies to accurately estimate the vehicle EFs.

Under the previous perspective, this PhD thesis aims to fulfill some of these scientific gaps. Alternative methodologies to assess road traffic emissions are developed and implemented to assess road traffic emissions in Ho Chi Minh City, Vietnam.

REFERENCES

Autonews., 2008. Global market data book.
http://www.autonews.com/

Colvile, R., Hutchinson, E., Mindell, J., Warren, R., 2001. The transport sector as a source of air pollution. Atmospheric Environment 35, 1537-1565.

Cheng, Y., Lee, S., Ho, K., Louie, P., 2006. On-road particulate matter (PM2.5) and gaseous emissions in the Shing Mun Tunnel, Hong Kong. Atmospheric Environment 40, 4235-4245.

Demerjian, K., 2000. A review of national monitoring networks in North America. Atmospheric Environment 34, 1861-1884.

EEA, European Environmental Agency., 2007. EMEP/CORINAIR emission Inventory Guidebook 2007.

Fenger, J., 1999. Urban air quality. Atmospheric Environment 33, 4877-4900.

Guo, H., Wang, T., Simpson, I., Blake, D., Yu, X., Kwok, Y., Li, Y., 2004. Source contributions to ambient VOCs and CO at a rural site in eastern China. Atmospheric Environment 38, 4551-4560.

Li, X., Liu, C., Leung, D., Lam, K., 2006. Recent progress in CFD modelling of wind field and pollutant transport in street canyons. Atmospheric Environment 40, 5640–5658.

McCarthy, M., Hafner, H., Chinkin, L., Charrier, J., 2007. Temporal variability of selected air toxics in the United States. Atmospheric Environment 41, 7180–7194.

Mellios, G., Van Aalst, R., Samaras, Z., 2006. Validation of road traffic urban emission inventories by means of concentration data measured at air quality monitoring stations in Europe. Atmospheric Environment 40, 7362–7377.

Molina, M., Molina, L., 2004. Megacities and atmospheric pollution. Air and Waste Management Association 54, 644–680.

Palmgren, F., Berkowicz, R., Ziv, A., Hertel, O., 1999. Actual car fleet emissions estimated from urban air quality measurements and street pollution models. Science of the Total Environment 235, 101-109.

Parrish, D., 2006. Critical evaluation of US on-road vehicle emission inventories. Atmospheric Environment 40 2006, 2288–2300.

Ropkins, K., Beebe, J., Li, H., Daham, B., Tate, J., Bell, M., Andrews, G., 2009. Real-world vehicle exhaust emissions monitoring: review and critical discussion. Critical Reviews in Environmental Science and Technology 39, 79-152.

US-EPA, Environmental Protection Agency., 2009.
http://www.epa.gov/

Vardoulakis, S., Fisher, B.E.A., Pericleous, K., Gonzalez-Flesca, N., 2003. Modelling air quality in street canyons: A review. Atmospheric Environment 37, 155–182

Watson, J., Zhu, T., Chow, J., Engelbrecht, J., Fujita, E., Wilson, W., 2002. Receptor modeling application framework for particle source apportionment. Chemosphere 49, 1093-1136.

World Health Organization, WHO., 2005. Health effects of transport-related air pollution.

WHO, World Health Organization., 2009
http://www.who.int/en/

Zarate, E., Belalcazar, L., Clappier, A., Manzi, V., Van den Bergh, H., 2007. Air quality modelling over Bogota, Colombia: Combined techniques to estimate and evaluate emission inventories. Atmospheric Environment 41, 6302–6318.

CHAPTER 3

VOCs AND OTHER POLLUTANTS ASSOCIATED TO MOTORCYCLES IN THE ATMOSPHERE OF HO CHI MINH CITY, VIETNAM

ABSTRACT

Motorcycles account up to 75% of the fleet in many Asian cities including Ho Chi Minh City (HCMC), Vietnam. Different studies show that motorcycles are one of the main sources of pollution in these cities but most of the pollutants produced by this mode of transport are not well identified. In this study we monitored online nineteen $C_2 - C_6$ VOCs, $PM_{2.5}$ and NO, together with traffic volumes during two months in the Ba Thang Hai (BTH) street, a busy road of HCMC. The traffic assessment shows that the BTH street is highly transited by motorcycles (95%), the remaining 5% are cars, trucks and buses. This study aims to assess the roadside levels of pollution for such traffic conditions and to identify their sources.

Results show that benzene and $PM_{2.5}$ levels are well above the World Heath Organization guidelines. N-hexane, i-pentane, and 3-methylpentane are the three most abundant species and account for about 60% of the total VOCs. A comparison with available studies demonstrates that selected VOC levels are the highest roadside levels reported in Asia. Further, the Maximum Incremental Reactivity (MIR) was used to compute the Ozone Formation Potential (OFP) of the measured VOCs. Trans-2-pentene, i-pentane, propene and n-hexane have the highest OFP, these four VOCs have the potential to generate 44% of the total ozone produced by all the VOCs included in this study.

A Principal Component Analysis (PCA) together with the analysis of daily variations of traffic volume and pollutants were used to identify their sources. Results show that all the VOCs (except isoprene) are produced by gasoline powered vehicles

(motorcycles), whereas NO is associated to diesel powered vehicles (buses and trucks). This analysis also reveals that road traffic is not an important direct source of $PM_{2.5}$ in HCMC.

Keywords: Volatile organic compounds, online gas chromatography, roadside measurements, receptor modeling

3.1. INTRODUCTION

The motorcycle fleet is rapidly increasing in many countries of the world. Motorcycles offer several advantages over cars and the public transport: lower purchase cost, cheaper to run, easer to park, and more flexible in traffic. For this reasons motorcycles account up to 75% of the fleet in Asia. In Vietnam, Thailand and Indonesia each have motorcycle sales of more than one million units per year (GTZ, 2002).

Different studies have shown that motorcycles are one of the main sources of pollutants in many Asian cities (Cai et Xie, 2007). Most of the air quality studies developed in such countries have been focused on the study of particles and ozone, but few studies have been conducted to study Volatile Organic Compounds (VOCs). VOCs are important air pollutants not only because they play a key role in atmospheric chemistry but also because some VOCs have toxic health effects.

Very few studies have been conducted in Vietnam to study VOCs. Truc and Oanh (2007) used charcoal tubes to monitor BTEX roadside levels in Hanoi, North Vietnam; a Principal Component Analysis (PCA) receptor model was also used to reveal the relationship between pollutant levels and traffic. Up to our knowledge, no long-term study has been published on the VOC characterization in Vietnam. Most of the studies conducted only include criteria pollutants, in some cases toxic pollutants have been measured during relatively short periods of time.

In this study we continuously measured roadside levels of 19 VOCs species in the range C2 – C6, together with $PM_{2.5}$ and NO. Measurements were taken during the dry season of 2007 in Ho Chi Minh City (HCMC), the largest city of Vietnam. The measurement campaign took place in a street highly transited by motorcycles. VOC

concentrations were measured with an online gas chromatograph; the other pollutants were also measured using standard automatic devices. The main objectives of this study are: to assess the roadside levels of VOCs and the other pollutants in HCMC and to identify their sources by means of a Principal Components Analysis (PCA) receptor model and the analysis of the daily variations of traffic flow and pollutants.

3.2. METHODOLOGY

3.2.1. Sampling site

The measurement campaign took place in the Ba Thang Hai (BTH) street, close to the centre of Ho Chi Minh City (HCMC) Vietnam. HCMC is the largest city of Vietnam, there are about 7 million inhabitants living in the city. HCMC has a typical tropical climate with two seasons, a rainy season from May to December, and a dry season from January to April. Ambient temperature is high especially in the dry season, where the average temperature ranges between 26 to 28 °C.

Figure 3.1. Left: Overview of the measurement site (1: mobile station; 2: traffic video recording (Adapted from Google Earth Inc). Right: A picture of the Ba Thang Hai street

The measurement site was located in the BTH street close to the intersection with the CMT8 street. The BTH street is a highly congested two-way street, it has three lanes each way. As it is common in HCMC, the BTH street is highly transited by motorcycles, followed by cars, busses and trucks. At the measurement site, the street is 24 m wide. There are sidewalks on both sides of the street of about 4 m wide. The buildings surrounding the street are in average 14 m high; more details about the site and the measuring campaign are shown in figure 3.1.

3.2.2. VOCs and other pollutants monitoring

An online Syntech Spectras 955 gas chromatograph (GC) was used for the measurements (Syntech, 2006). C2 – C6 VOCs are preconcentrated on carbosieve SIII at 5 °C, desorbed thermally and separated on a combination of two columns, a capillary film column and a capillary PLOT column. Analysis is done by PID and FID detectors. The instrument detection limits are: 0.1 ppbv for aromatics; 0.1 ppbv for alkenes from C3; 0.5 ppbv for alkanes between C3 and C4 and 0.1 ppbv for alkanes from C5.

In this study, continuous measurements in 30 minute intervals of 17 VOCs were recorded during the campaign. VOCs and the other pollutants measurements started on January 9 2007 at 12:30 pm and ended on March 11 2007 at 9:30 am. The GC was installed inside a mobile air quality monitoring station, the mobile station was placed in the street beside the east sidewalk (see figure 3.1). $PM_{2.5}$ were monitored with an Environment S.A Ambient Air Suspended Particles Analyzer (model MP101M, Beta Gauge); and NO with an Environment S.A Chemiluminesent Nitrogen Oxides Analyzer (model AC32M). Air samples were continuously collected from the top of the mobile station at 2.5 m.

The GC was calibrated according to Syntech recommendations; one complete full dynamic calibration was performed during the two months of campaign, a span check was also performed every week. A standard certified calibration gas at 10 ppm for 16 VOCs species and 100 ppm for propane was purchased (see the list of species in table 1). The calibration gas was diluted with zero air (quality 5.0) to prepare five calibration spans; the dilutions were prepared using two Bronkhorst digital mass flow controllers (0 – 500 ml air min-1: accuracy ± 0.5%; 0 – 1 ml N2 min-1: accuracy ± 2%). In addition to the 17 VOCs species, two additional species (1,3–butadiene and isoprene) were also present in the calibration bottle at around 10 ppm. However,

since this two species were not certified by the manufacturer of the calibration bottle, the GC could not be calibrated for quantitative measurement of these components. Nevertheless, the source identification requires only normalized concentrations and then these substances can still be used for the identification.

3.2.3. Traffic counts

Traffic flow in the BTH street was continuously recorded with a video camera (see figure 3.1). The Videos were saved onto a hard disk and periodically burned to DVDs. Traffic volumes were counted manually after the measurement campaign. The fleet was divided in four vehicle categories for the counting: motorcycles, cars, trucks and busses. Traffic volumes in HCMC present two peaks, one in the morning and one in the afternoon; additionally, in HCMC there are two heavy traffic (trucks) restrictions, one in the morning from 7 to 9 am, and one in the evening from 17 to 20 pm. Due to the huge amount of data, we decided to count 30 non consecutive days out of the 60 days available, traffic volumes were also counted from 10 am to 22 pm, we chose this period to cover one of the two peaks of traffic and one of the traffic restriction of the day.

3.3. RESULTS AND DISCUSSION

3.3.1 Fleet distribution

About 6.5 million motorcycles, 330 thousand cars, 11 thousand trucks and 20 thousand buses were counted during the 30 days selected for the traffic volumes assessment. Due to local existing regulations almost all of these motorcycles use four-stroke engines.

Figure 3.2 shows the fleet distribution observed in the BTH street. As can be seen, motorcycles represent almost 95% of the total fleet passing through the site. Figure 3.2 also shows the fleet distribution reported in other streets of HCMC, this distribution is based on counts developed for the HOUTRANS study (HOUTRANS, 2004), the counts were conducted to grasp the traffic volumes at major roads in HCMC and to calibrate traffic forecast models; 24 hours counts were developed from November 19 to December 2 (2001) in 37 major roads of HCMC. As can be seen in

figure 3.2, the fleet distribution in the BTH street is similar to that obtained from the HOUTRANS study, this indicates that the results of the present study may be representative of the general situation in the city. There is a difference in the percentage of trucks circulating though the BTH street with respect to these found in the other streets of the city, this difference can be due to the periods of the day selected for the counting, the distribution reported in the other streets is based on 24 hours counts, whereas the fleet distribution reported here for the BTH street is based on counts developed mostly during the day (from 10 am to 22 pm). Due to the heavy traffic restrictions, during night time there are trucks circulating in the street and these trucks surely modify the fleet distribution.
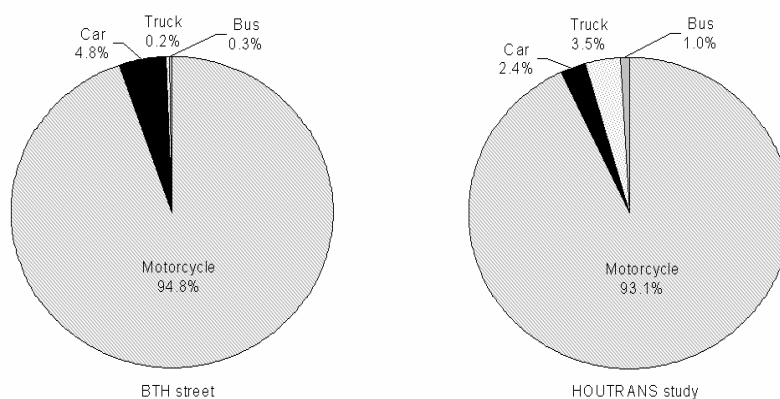
Figure 3.2. Fleet distribution in the BTH street (this study) and in other street of HCMC (HOUTRANS, 2004)

3.3.2 Roadside concentrations

In this study, about 2600 30-minute average concentrations were collected during the two months of campaign for most of the VOC species, and about 1500 for $PM_{2.5}$, and NO. In the case of n-propane about 500 data and about 1600 data for n-butane are available This lower number of data for these two VOCs is due to the following reason: during the measurement campaign, LPG gas (39% n-propane; 30% n-butane; 30% i-butane; and 1% other VOCs) was used as a tracer for a separate study. In that study, LPG was emitted during some periods of time in the street, thus, n-propane and n-butane data corresponding to these periods of time were removed from the data set used for this study (note that i-butane was not included in this study). Results of that tracer experiment will be presented in a separated paper.

Table 3.1. Statistics and properties of VOCs and other pollutants measured in the BTH street (concentrations are in ppbv otherwise specified)

Compound[a]	Mean	SD[b]	MAX[c]	OFP[d] $\mu g\, O_3\, m^{-3}$	% of Total 17 VOCs	% of Total OFP
n-Propane	3.7	3.8	32.5	3.1	1	0
Propene	19.5	14.3	64.7	320.9	5	**13**
n-Butane	22.6	19.7	115.7	55.6	6	2
Trans-2-Butene	5.2	3.8	23.9	121.4	1	5
1-Butene	4.0	3.0	13.4	83.9	1	3
Cis-2-butene	5.2	3.9	63.4	121.2	1	5
i-Pentane	80.3	55.5	347.3	332.0	**23**	**14**
n-Pentane	21.8	15.3	88.4	67.9	6	3
Trans-2-Pentene	16.4	10.3	46.9	421.7	5	**17**
1-Pentene	4.6	2.9	23.5	83.3	1	3
2-methyl-2-butene	3.8	2.7	13.7	71.2	1	3
Cis-2-Pentene	4.2	2.8	17.4	108.7	1	4
2,3-Dimethylbutane	8.6	6.2	35.1	32.9	2	1
2-Methylpentane	7.7	5.4	55.6	41.1	2	2
3-Methylpentane	43.5	30.0	127.8	233.3	**12**	10
n-Hexane	91.0	58.9	302.8	319.1	**26**	**13**
Benzene	14.2	9.6	51.3	19.4	4	1
Total	356			2437		
NO	92.5	73.8	427.0			
$PM_{2.5}$ ($\mu g\, m^{-3}$)	72.1	33.7	392.1			

[a] 1,3-butadiene and isoprene are not presented in this table, see section 3.2.2 for details

[b] SD: Standard deviation

[c] MAX: maximum value registered during the campaign

[d] OFP: Ozone Formation Potential. OFP = MEAN ($\mu g\, m^{-3}$) x MIR (MIR; Maximum Incremental Reactivity: g O_3 / g VOC. Carter, 1997)

Table 3.1 summarizes statistics and properties of the 17 VOCs included in this study together with other pollutants monitored. As can be seen, n-hexane, i-pentane, and 3-methylpentane are the most abundant of the 17 VOCs, these three species account near to 60% of the total VOCs measured. The ozone formation potential is the capacity of a VOC to produce ozone. The amount of ozone produced not only

depends on the amount of VOC emitted but also on its reactivity. Carter (1997) developed methods for ranking photochemical ozone formation reactivities of VOCs, one of the most widely used method is the Maximum Incremental Reactivity (MIR). Using this scale, the ozone formation potential (OFP) is calculated multiplying the average concentration registered by the MIR of each VOC species. The OFP calculated are presented in table 3.1 for each VOC together with its contribution in percentage to the total potential ozone produced. As can be seen, trans-2-pentene, i-pentane, and n-hexane have the potential to generate 44% of the total ozone produced by the VOCs included in this study.

Air quality standards for the individual VOC species have not been defined yet; the World Health Organization (WHO) has defined some guidelines for some individual VOCs based on effects other than cancer and odor nuisance (Sirvastava et al., 2006). Among these substances is benzene, a recognized carcinogenic pollutant. The average benzene concentration registered in the BTH street during the two months of campaign was 14.2 ppbv (see table 3.1), this concentration exceeds the annual mean WHO guideline of 6 ppbv. A short-term (30 minutes) air quality standard of 9 ppbv for benzene has been established in Texas USA (Zhao et al., 2004), benzene levels in the BTH street exceeded this environmental standard 63% of the time. WHO has also defined guidelines for $PM_{2.5}$ (WHO, 2005), the average $PM_{2.5}$ concentration in the BTH street was 72.1 µg m^{-3}, this value largely exceeds the annual mean WHO guideline of 10 µg m-^{3}.

Roadside VOC measurements are scarce even in developed countries, just few roadside studies have been developed in Asia and only selected VOCs have been measured. Table 3.2 presents a comparison of selected VOC levels in the BTH street with available roadside studies reported for other Asian cities. As can be seen, i-pentane and n-hexane levels in the BTH street are the highest reported in Asia. The levels of benzene in Hanoi are higher than levels found in the BTH street. It should be noted that the roadside benzene levels reported for Hanoi correspond to these measured in the polluted busy roadsides, mainly at peak hours, and in the most polluted months of the year, which are likely the highest observed in that city (Truc and Oanh, 2007)

Table 3.2. Comparison of selected VOCs levels in the BTH street with other roadside studies

VOCs (ppbv)	BTH street Mean	Changchun[a] Mean	Karachi[b] Mean	Hong Kong[c] Mean	Hanoi (TC)[d] GM	Hanoi (NT)[d] GM
i-pentane	80.3	14.7	74			
2-Methylpentane	7.7	6.1	39			
n-Hexane	91.0	1.7	71	4.4		
Benzene	14.2	11.9	19.7	8.2	20	40

[a] Liu et al., 2000.

[b] Barletta et al., 2002.

[c] Chan et al., 2002.

[d] Truc and Oanh., 2007. TC: Truong Chinh street. NT: Nguyen Trai street (GM: geometric mean)

3.3.3 Daily variations of traffic and pollutants

Figure 3.3 presents the daily variations of the different vehicle categories (figure 3.3A), $PM_{2.5}$, and NO (figure 3.3B), and the 19 VOCs species considered in this study (figure 3.3C). These plots were obtained by calculating the half and hour mean for each of the pollutants considered in this study from all the data gathered during the campaign. In the case of Figure 3.3B and 3.3C, data were normalized before computing the 30 minutes mean values, normalization was done dividing each individual concentration of the pollutant by its own mean.

As can be seen from figure 3.3A, the afternoon rush hour for motorcycles and buses is between 17 - 18 pm; at this afternoon peak, on average, nearly 12 thousand motorcycles circulate through the street in just 30 minutes. This number is significantly higher in comparison to the 380 cars and 45 buses that circulate in the street during the same time. As mentioned before, heavy traffic is banned in HCMC from 17 to 20 pm. It can be seen in figure 3.3A that the average number of trucks drops to zero during this period of time; nevertheless, during the counting it was found that usually one or two trucks were evading the restriction. Figure 3.3A also shows that just after the heavy traffic restriction, the number of trucks increases rapidly to reach its highest peak between 21 - 22 pm.
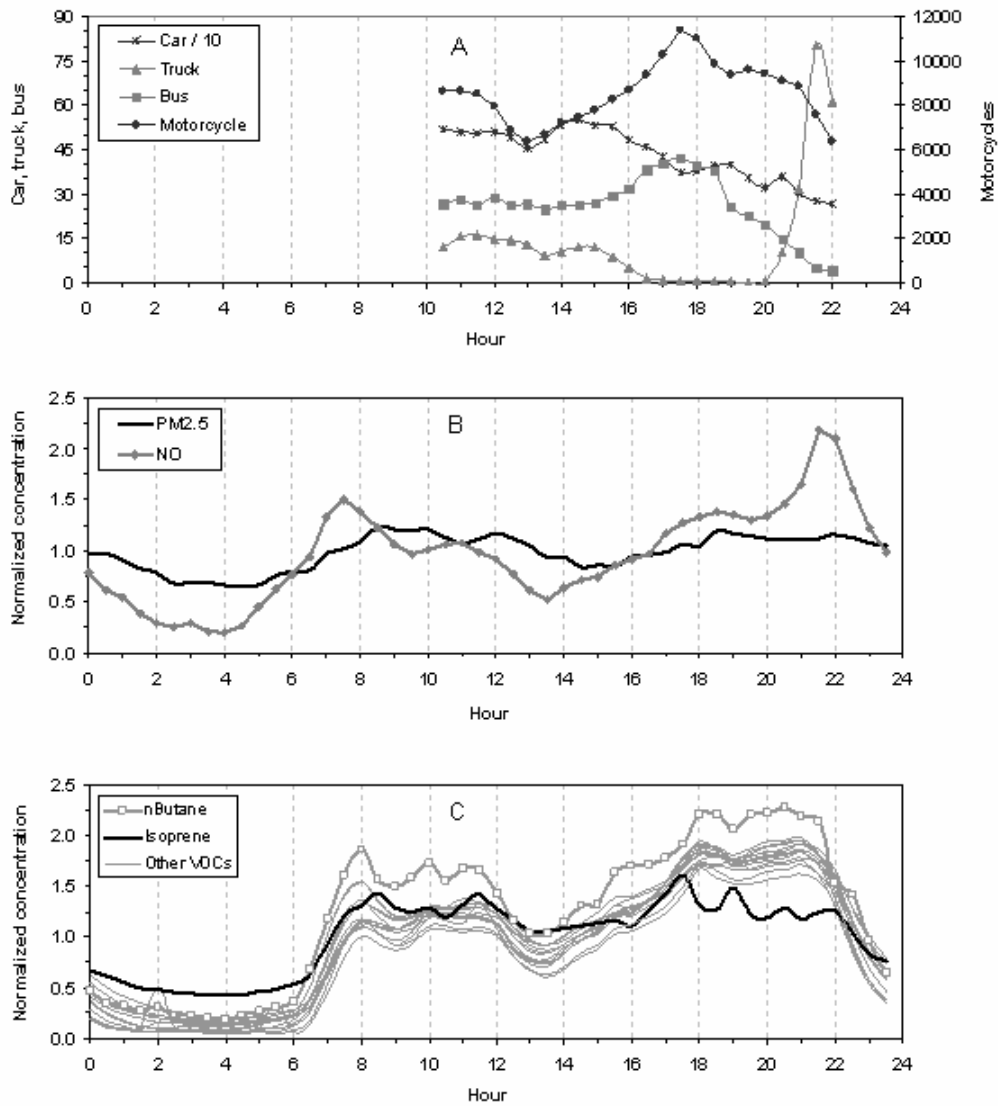
Figure 3.3. Daily 30 minute variations of traffic, VOCs and other pollutants. A: Mean vehicle volumes in the BTH street (CAR / 10: the number of cars is divided by 10). B: Mean normalized concentrations of $PM_{2.5}$ and NO. C: Mean normalized concentrations of the 19 VOCs considered in this study.

The 30 minute daily variations of NO are presented in figure 3.3B. Diurnal variations of NO present two peaks, one in the morning and one in the evening; at night time (from 2 to 5 am) concentrations of this substance go down to values close to zero, although it is not shown in figure 3.3A, at this time traffic volumes are also very low, this indicates that NO levels in the BTH street are mainly related to road traffic emissions. Daily variations of NO are similar to the daily variations of trucks and buses, the highest peak of NO occurs between 21 - 22 pm. This peak coincides with the truck peak observed at the same time in figure 3.3A. Trucks and buses in HCMC are mostly diesel powered vehicles, which are recognized to be an important source of NO. Thus, the NO levels in the BTH street can also be closely associated to diesel powered vehicles.

Daily variations of $PM_{2.5}$ are also presented in figure 3.3B, as can be seen, levels of $PM_{2.5}$ fluctuate close to the normalized mean value (the mean normalized value is equal to 1.0), this indicates that there is an important background of this pollutant present in the street. Levels of $PM_{2.5}$ do not seem to follow the traffic flow variations. Hien et al (2001) used a PCA model to identify and apportion the sources of PM_2 in HCMC, it was found that 25% of PM_2 was produced by chemical reactions, it was also found that 35% of these particles were produced by soil dust, road dust, and biomass burning; only 17% of the PM_2 was attributed to direct vehicles emissions; the resting 23% was attributed to industry and coal combustion. Results of Hien's study agree with results presented here, then, we can conclude that road traffic is not an important source of $PM_{2.5}$ in HCMC.

Figure 3.3C presents the mean normalized diurnal variations of the 19 VOCs species include in this study. Most of the VOC concentrations follow the same trend. VOC concentrations show two peaks, one in the morning and one in the evening, the evening peak is subdivided into two peaks, each one matches with the evening peaks of motorcycles, buses and trucks observed in figure 3.3A; additionally, VOC levels increase during the day and decrease to values close to zero at night. This behavior indicates that VOCs can be associated to a common source, the direct roadside traffic emissions. n-butane also presents the same behavior followed by the other VOCs but the n-butane line is above the other VOCs, this is because the levels of this VOC frequently exceed its mean value. Only isoprene exhibits a slight different behavior, especially during the afternoon. Isoprene is considered to be emitted by biogenic sources, this may explain this behavior. Nevertheless the variations of

isoprene during the morning seems to be affected by traffic variations (figure 3.3C), road traffic has also been identified as a source of isoprene (Barletta et al., 2005).

3.3.4 Source identification

A principal Components Analysis (PCA) is used to identify the pollutant sources and to verify the source identification presented in section 3.3. The statistical package SPSS version 13.0 was used for the PCA. The details of the PCA can be found elsewhere (Thurston and Spengler, 1985; Miller et al., 2002; Guo et al., 2004). Briefly, PCA is a multivariate receptor model that can be used to identify a reduced number of independent factors from a large number of variables, each factor corresponds to a source or a group of sources which can be associated to different pollutants, the association is achieved by using the loadings produced by the PCA analysis.

In this study, the 19 VOCs, $PM_{2.5}$, and NO are used for the PCA. Table 3.3 presents the PCA results. To simplify the interpretation of the results, a Varimax rotation with Kaiser normalization is applied; only factor loadings greater that 0.3 are presented. Three factors with eigenvalues over 1.0 were extracted.

Factor number 1 (F1) has high loadings for all the VOCs except isoprene. VOCs like n-butane, i-pentane, n-pentane and benzene have been associated to gasoline vehicle emissions and gasoline evaporation in the literature (Guo et al., 2007). Thus, F1 most likely corresponds to gasoline powered vehicles, which in HCMC are mainly motorcycles.

Factor number 2 (F2) has a high loading for isoprene, and, as it was mentioned before, isoprene is associated to biogenic sources. Nevertheless, this VOC has been also attributed to road traffic. NO has also a high loading in F2, NO is attributed to diesel powered vehicle emissions. Therefore, F2 may include biogenic and diesel emissions.

Table 3.3. PCA results for 19 VOCs, $PM_{2.5}$ and NO[a]

	F1	F2	F3
n-Propane	0.31	-0.43	0.51
Propene	0.92		
n-Butane	0.90		
Trans-2-Butene	0.96		
1-Butene	0.97		
Cis-2-butene	0.95		
i-Pentane	0.97		
n-Pentane	0.96		
1,3 Butadiene	0.93		
Trans-2-Pentene	0.95		
1-Pentene	0.94		
2-methyl-2-butene	0.96		
Cis-2-Pentene	0.97		
2,3-Dimethylbutane	0.95		
2-Methylpentane	0.81	0.53	
3-Methylpentane	0.94		
n-Hexane	0.94		
Isoprene		0.77	
Benzene	0.91		
NO		0.80	
$PM_{2.5}$ ($\mu g\ m^{-3}$)			- 0.83
Initial eigenvalues	**15.2**	**1.9**	**1.1**
% of variance	**72.3**	**9.1**	**5.2**

[a] Extraction method: PCA. Rotation method: Varimax with Kaiser normalization

As can be seen in table 3.3, n-Propane has a moderate loading of 0.51 in factor number 3 (F3), n-propane is a good indicator of LPG emissions. There is also a high loading of $PM_{2.5}$ in F3 and there is no association of this pollutant to F1 and F2. As mentioned before, fine particles in HCMC have been attributed to other sources than traffic, this PCA analysis confirms that road traffic is not an important direct source of $PM_{2.5}$.

3.4. CONCLUSIONS

For a first time in Vietnam, online roadside measurements of VOCs were developed. Roadside levels of the pollutants measured are above the available WHO guidelines; moreover, concentrations of selected VOCs are the highest reported in Asia. This study also let the identification of the main VOCs ozone precursors; it was also possible to identify VOCs emitted by road traffic in the city. An analysis of daily variations of traffic and pollutant, together with a Principal Components Analysis revealed that all the VOCs included in this study except isoprene are emitted by gasoline powered vehicles (mostly motorcycles). This study confirmed the results of a previous study, it was confirmed that road traffic in HCMC is not an important source of fine particles.

Results of this work indicate that HCMC is highly affected by air pollution. It is necessary to adopt policies to improve air quality in the city, these policies should include the implementation of strategies for a better traffic management like the development of better public transport systems and the reduction of the motorcycles fleet in the city; other strategies are also necessary like the improvement of fuel quality and vehicle technologies.

ACKNOWLEDGMENTS

REFERENCES

Barletta B, Meinardi S, Simpson I, Khwaja H, Blake D, Rowland S. Mixing ratios of volatile organic compounds (VOCs) in the atmosphere of Karachi, Pakistan. Atmos Environ 2002; 36: 3429–3443.

Barletta B, Meinardi S, Sherwood R, Chan C, Wang X, Zou S, Chan L, Blake D. Volatile organic compounds in 43 Chinese cities. Atmos Environ 2005 ; 39 : 5979-5990.

Cai H, Xie S. Estimation of vehicular emission inventories in China from 1980 to 2005. Atmos Environ 2007; 4:, 8963-8979.

Carter W.P.L. Maximum Incremental Reactivity Excel Spreadsheet. http://pah.cert.ucr.edu/~carter/rcttab.htm 1997

Chan C, Chan L, Wang X, Liu Y, Lee S, Zou S, Sheng S, Fu J. Volatile organic compounds in roadside microenvironments of metropolitan Hong Kong. Atmos Environ 2002; 36: 2039–2047.

Guo H, Wang T, Louie P.K.K. Source apportionment of ambient non-methane hydrocarbons in Hong Kong: application of a principal component analysis (PCA)/absolute principal component scores (APCS) receptor model. Environ Pollut 2004; 129: 489–498.

Guo H, So K, Simpson J, Barletta B, Meinardi S, Blake D. C1-C8 volatile organic compounds in the atmosphere of Hong Kong: Overview of atmospheric processing and source apportionment. Atmos Environ 2007; 41: 1456-1472.

GTZ. Sustainable transport: A source book for policy-makers in developing cities; module 4C: two and three-wheelers. 2002.

Hien P, Binh N, Truong Y, Ngo N, Sieu L. Comparative receptor modelling study of TSP, PM_2 and PM_{2-10} in Ho Chi Minh City. Atmos Environ 2001; 35: 2669-2678.

HOUTRANS. The study on urban transport master plan and feasibility study in Ho Chi Minh metropolitan area. 2002; Volume 6, No 1 traffic and transport surveys. ALMEC Corporation.

Liu C, Xu Z, Du Y, Guo H. Analyses of volatile organic compounds concentrations and variation trends in the air of Changchun, the northeast of China. Atmos Environ 2000; 34: 4459-4466.

Miller S, Anderson M, Daly E, Milford J. Source apportionment of exposures to volatile organic compounds I. Evaluation of receptor models using simulated exposure data. Atmos Environ 2002; 36: 3629–3641.

Sirvastava A, Joseph A, Devotta S. Volatile organic compounds in ambient air of Mumbai—India. Atmos Environ 2006; 40: 892–903.

Syntech. Manual for the Syntech Spectras GC955 series 400, 600, 800, single and double. 2006.

Thurston G, Spengler J. A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston. Atmos Environ 1985; 19: 9–25.

Truc V, Oanh N. Roadside BTEX and other gaseous air pollutants in relation to emission sources. Atmos Environ 2007; 41: 7685–7697.

World Health Organization (WHO). WHO air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. Global update 2005.

Zhao L, Wang X, He Q, Wang H, Sheng G, Chan L, Fu J, Blake D. Exposure to hazardous volatile organic compounds, PM10 and CO while walking along streets in urban Guangzhou, China. Atmos Environ 2004; 38: 6177–6184.

CHAPTER 4

ESTIMATION OF ROAD TRAFFIC EMISSION FACTORS FROM A LONG TERM TRACER STUDY IN HO CHI MINH CITY (VIETNAM)

Abstract

Road traffic emissions, one of the largest source categories in megacity inventories, are highly uncertain. It is essential to develop methodologies to reduce these uncertainties to manage air quality more effectively. In this paper, we propose a methodology to estimate road traffic emission factors (EFs) from a tracer experiment and from roadside pollutants measurements. We emitted continuously during about 300 non-consecutive hours a passive tracer from a finite line source placed on one site of an urban street. At the same time, we measured continuously the resulting tracer concentrations at the other side of the street with a portable on-line gas chromatograph. We used n-propane contained in commercial liquid petroleum gas (LPG) as a passive tracer. Propane offers several advantages to traditional tracers (SF_6, N_2O, CFCs): low price, easily available, non-reactive, negligible global warming potential, and easy to detect with commercial on-line gas chromatographs.

The tracer experiment was carried out from January to March 2007 in a busy street of Ho Chi Minh City (Vietnam). Traffic volume, weather information and pollutant concentrations were also measured at the measurement site. We used the results of the tracer experiment to calculate the dilution factors and afterwards we used these dilution factors, the traffic counts and the pollutant concentrations to estimate the EFs. The proposed method assumes that the finite emission line represents the emission produced by traffic in the full area of the street and therefore there is an error associated to this assumption. We use the Computational Fluids Dynamics model (CFD) MISKAM to calculate this error and to correct the HCMC EFs. EFs for 15 Volatile Organic Compounds (VOCs) and NO are reported here. A comparison

with available studies reveals that most of the EFs estimated here are within the range of EFs reported in other studies.

Keywords: Real-world emissions, street canyon, on-line gas chromatography, roadside measurements, tracer experiment

4.1. INTRODUCTION

Different techniques exist to identify the sources of pollution in cities, among these techniques are emission inventories. Although emission inventories are an essential tool for managing and regulating pollution, large uncertainties in emission rates, temporal cycles, spatial distributions and source identification often confound the development of cost-effective control strategies (Molina and Molina, 2004). Emission inventories apply an emission factor that represents the mass of emission per unit of activity times an activity factor. Emission factors and activity levels are highly uncertain for vehicles, one of the largest source categories in megacity inventories. It is essential to reduce these uncertainties to manage air quality more effectively.

Two approaches exist to estimate traffic emission factors. In the traditional approach (bottom-up) emission factors are determined by measuring directly the emission at the exhaust of the vehicle. In the last years, a different approach has been developed (top down), this approach is based on the indirect estimation of the emission factors. Both approaches offer several advantages and face limitations, and thus, both complement each other.

Different methodologies can be considered as top down techniques, the most widely used are tunnel studies and inverse modeling. Tunnel studies are an interesting alternative to estimate emission factors. However, it is not always possible to find a tunnel close or inside a city where the emissions are produced and which would represent in a better way the real-world urban conditions. Inverse modeling has been used to estimate emission factors in different cities of the world (Palmgren et al., 1999: Olsece et al., 2001; Gramotnev et al., 2002; Ketzel et al., 2003; Kawashima et al., 2006). The advantage of inverse modeling is that it is possible to estimate the emissions under real-world urban conditions. On the other hand, since the method uses an air quality model to estimate the emission factors, the accuracy of the

estimated emission factors depends on the ability of the model to reproduce the dispersion of the pollutants. Studies have shown that one model might perform better than an alternative model in one study but results may be reversed in a different scenario (Holmes and Morawska, 2006). Inter-comparison studies and evaluation of models show discrepancies in the modeling results (Vardoulakis et al., 2003).

Alternative top down techniques have been developed to estimate and evaluate traffic emissions. Claiborn et al. (1995) estimated PM_{10} emission factors using a tracer technique. A known amount of an inert tracer was released. Samples downwind from the source were collected and analyzed at the laboratory. Results from this tracer experiment together with measured PM_{10} concentrations, and available meteorological information and traffic volumes were used to estimate PM_{10} emission factors. Results of this tracer experiment showed that the use of a tracer in a finite line source provide a tool for improving the existing emission inventories.

Hueglin et al. (2006) derived relative traffic emission factors in a motorway from long term air quality data. The proposed methodology uses dilution factors, roadside and background concentrations of pollutants to calculate relative traffic emission factors. Since the dispersion factors are unknown, they were estimated in relative terms by assuming unity emission factors for a reference year. Results from that study were used to estimate long term trends of traffic emission factors but absolute emission factors couldn't be estimated.

In this study, we designed and tested a system to estimate dilution factors and traffic emissions from a tracer study. We continuously released a passive tracer on one side of an urban street and at the same time we measured the resulting concentrations on the other side of the street. We used these results to calculate the dilution factors and hence the traffic emission factors. The use of this technique offers two advantages. First, a large acquisition of data thanks to the on-line measurements. Second, the estimation of actual emission factors obtained at urban conditions from calculated dilution factors which are accurately known since we know the amount of tracer released.

4.2. METHODOLOGY

This study was carried out in Ho Chi Minh City (HCMC) Vietnam. HCMC is the largest city in Vietnam and one of the largest cities of the Southeast Asia. HCMC is located at sea level, it has a typical tropical climate with two seasons, a rainy season from May to December, and a dry season from January to April. Ambient mean temperatures are especially high in the dry season when they range between 26 to 28 °C.

The high demand for transport in the city, and all the advantages offered by motorcycles made them the main mode of transport in HCMC. In this city there are about 4 million motorcycles which are 78% of the total fleet of the city. In HCMC most of the motorcycles use 100 cc four-stroke engines. The rest of the fleet is composed of car buses and trucks. The particular conditions of the city and especially the limited funding make the use of a traditional methodology to estimate and evaluate the road traffic emissions difficult.

4.2.1. Measurement site

The tracer experiment was conducted in the Ba Thang Hai street (BTH, see figure 4.1), close to the centre of HCMC. BTH street is a highly congested two-way street, it has three lanes each way. As it is common in HCMC, the BTH street is highly transited by motorcycles, followed by cars, busses and trucks. At the measurement site, there are sidewalks on both sides of the street of about 4 m width; the street including the sidewalks is 24 m width. The BTH street is flanked at both sides by buildings of an average height of 14 m with occasional open spaces between them. The height to width ratio of the street canyon is 0.58. The distance between the two closer major intersections is about 350 m. According to these geometric characteristics, this street can be classified as a long symmetric avenue canyon (Vardoulakis et al., 2003). 28 m high trees stand at both sides of the street, they are very close one to the other and have dense crowns at the top (last 8 m).

Figure 4.1. Measurement site. 1: Emission liberation device over the Ba Thang Hai street. 2: mobile station. 3: traffic video recording. 4: Meteorological station. Adapted from Google earth.

4.2.2. Tracer selection and emission rate

In this study, n-propane contained in Liquid Petroleum Gas (LPG) was used as a passive tracer. Traditionally SF_6, N_2O and some CFCs have been used in tracer studies. Propane offers several advantages with respect to traditional tracers. Propane is commonly available, very cheap, non-reactive at the time scales of interest, it has negligible global warming potential, and it is easy to detect with commercial on-line gas chromatographs. Due to its characteristics, n-propane can be released continuously over long periods of time.

There is a background concentration of propane in the ambient air of the measurement site. Although it is not the main source of propane, road traffic is a source of this compound. Initial propane measurements at the site showed that roadside levels are around 4 ppbv. To avoid the effect of this propane background, we chose an emission rate of this compound to obtain levels well above this background concentration in the tracer study. A simple box model was used to

calculate the tracer emission rate needed to reach these levels. The calculations showed that a continuous propane emission rate of 0.21 Nm^3 h^{-1} (0.105 g s^{-1}) was enough to reach concentrations at street level of about 200 ppbv, which is 50 times above the typical background levels.

The composition of commercial LPG in HCMC is (% in volume): 39.1% n-propane; 30% n-butane; 30% i-butane; 0.2% ethane, 0.2% isopentane; 0.2% neopentane; and others 0.3%. Therefore we used 0.54 Nm^3 h^{-1} as the LPG emission rate. It should be noted that n-butane and i-butane couldn't be used as tracers since the background levels detected for these compounds went up to 100 ppbv. It should be also noted that concentrations of isopentane, neopentane and others in the LPG bottle are too low to affect typical roadside concentrations of these pollutants.

4.2.3. Experimental set up

The experimental set up consists of two parts. The first part is a tracer emission device and the second part is a portable on-line gas chromatograph used to measure the resulting tracer concentrations (points 1 and 2 in figure 4.1).

Tracer emission device

Figure 4.2 schematizes the tracer emission device. All the devices were installed inside a metallic cabinet to protect the equipment. Two LPG bottles were connected to a bottle switch. Once a bottle was empty the switch activated automatically the liberation from the other bottle, meanwhile the empty cylinder could be replaced by a full bottle without interrupting the tracer release.

The tracer emission rate was controlled by means of a LPG mass flow controller Red-y GSC smart series, 0 – 0.6 Nm^3 h^{-1}, accuracy 0.5% (MFC in figure 4.2). Emitted LPG was injected into a 5 cm internal diameter flexible hose. The LPG Lower Explosion Limit (LEL) is 2% Vol% (20 000 ppmv). To avoid the risk of explosion, it was necessary to dilute LPG with enough ambient air to stay at LPG concentrations well below the LEL. A centrifugal blower was used to generate the air needed for the dilution (Ebmpapst RG175 blower, flow 0 – 390 m^3 h^{-1}). The air flow produced by the blower depends on the pressure drop in the liberation system, in this case the air flow produced was 120 Nm^3 h^{-1}. Thus the mix LPG air was 4500 ppmv, or 21% of its LEL. As a security measure, a Evikon LPG E2606 LEL detector was continuously

monitoring the LPG concentration inside the hose. It was programmed to switch off the mass flow controller and stop the emission if the LPG concentration reached 50% of the LEL.
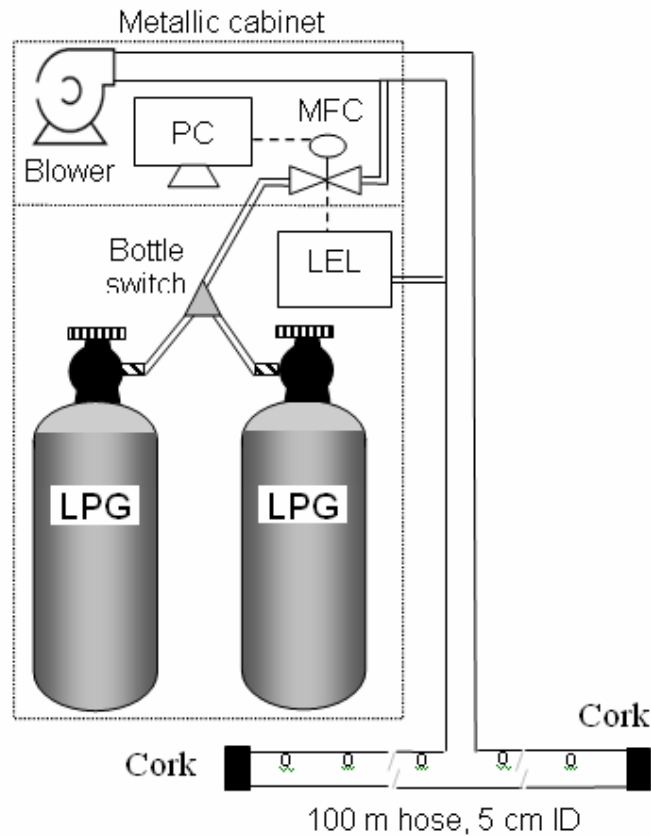

Figure 4.2. Tracer emission device

The LPG/air mix was injected into the middle of a 100 m hose which was located at ground level, between the west sidewalk curb and the lane, at 8 m from the axis of the street. The ends of the hose were closed with corks. In order to release the tracer uniformly along the street and approximate traffic emissions by a line source, holes of 1 mm diameter were perforated each meter over the whole hose. Air flow measurements at each hole showed that the flow was equivalent for all of them to a sufficient degree of accuracy. The tracer emission was thus uniformly distributed along the hose.

On-line gas chromatograph

The resulting tracer concentrations were monitored together with other VOCs at the other side of the street with a portable on-line $C_2 - C_6$ Syntech Spectras 955 gas chromatograph (GC) (Syntech, 2006). The GC was installed inside a mobile air quality monitoring station which was parked in the street, beside the east sidewalk at around 8 m from the axis of the street. The analysis was done with PID and FID detectors. $C_2 - C_6$ VOCs were preconcentrated on carbosieve SIII at 5 °C, desorbed thermally and separated on a combination of two columns, a capillary film column and a capillary PLOT column. The instrument detection limits are: 0.1 ppbv for aromatics; 0.1 ppbv for alkenes from C3; 0.5 ppbv for alkanes between C3 and C4 and 0.1 ppbv for alkanes from C5.

The GC was calibrated according to Syntech recommendations; one complete full dynamic calibration was performed during the campaign. A span check was performed every week. A standard certified calibration gas at 10 ppmv for 16 VOCs species (including n-butane) and 100 ppmv for propane was used. The calibration gas was diluted with zero air (quality N5.0) to prepare five calibration spans; the dilutions were prepared using two Bronkhorst digital mass flow controllers (0 – 500 mL air min^{-1}: accuracy ± 0.5%; 0 – 1 ml N_2 min^{-1}: accuracy ± 2%).

The tracer experiment was carried out during the dry season from January to March 2007. LPG was released during 25 non consecutive days starting at 10:00 and finished at 22:00. 30-minute composite samples were analyzed continuously with the on-line GC for propane, butane and 15 additional VOCs (table 4.1). $PM_{2.5}$ were monitored with an Environment S.A Ambient Air Suspended Particles Analyzer (model MP101M, Beta Gauge); and NO with an Environment S.A Chemiluminesent Nitrogen Oxides Analyzer (model AC32M). Air samples were continuously collected from the top of the mobile station at a height of 2.5 m.

4.2.4. Estimation of traffic emission factors

Considering the dispersion of non-reactive or slowly reactive gases, the chemical transformations are neglected, the following expression relates pollutant concentrations at a certain location and traffic emissions (Palmgren et al., 1999):

$$C_i = F_i \, Q_i + C_{b,i} \qquad\qquad (1)$$

Where C_i is the concentration of a particular pollutant in the street at any time i (µg m^{-3} or ppbv after converting by using the respective pollutant density), Q_i is the total average emission of a pollutant (g m^{-1} s^{-1}), and F_i (s m^{-2}) is a function describing dispersion processes at any time i, F_i is also called dispersion factor. $C_{b,i}$ is the contribution to pollutant concentrations in the street from all other sources than the traffic in the street at any time i, that is the background concentration (µg m^{-3} or ppbv). Fi is a complex function of meteorology (wind direction, wind speed, mixing heights, etc.), on the geometry of the street (building height, street width, etc.), and on the traffic induced turbulence.

F_i has been typically calculated by means of a street canyon dispersion model (Palmgren et al., 1999; Olcese et al., 2001; Ketzel et al., 2003; Zarate et al., 2007). Although this methodology is an interesting alternative to estimate traffic emissions, most of the available dispersion models have been validated using experimental datasets from deep canyons (Vardoulakis et al., 2007), that is those for which the ratio between the average height of the buildings and the width of the street is bigger than one. It is not always possible to find these kinds of canyon geometries in a city and thus this technique can not always be used to estimate the traffic emissions. Also, since the accuracy of a street canyon dispersion model is often questionable, such results are often attached with uncertainties.

Here we propose to estimate the dispersion factor F_i needed in equation (1) from the tracer experiment. As mentioned before, propane background concentrations are negligible (expected propane concentrations are about 50 times bigger than the background concentration), and thus F_i can be obtained as follows:

$$F_i = C_{t,\,i} \,/\, 1.05 \times 10^{-3} \qquad\qquad (2)$$

Where C_t is the measured concentration of tracer (µg m^{-3} or g (x 10^{-6}) m^{-3}) at the time i. In this case, since the available VOCs concentrations are measured in 30-minutes intervals, i correspond to periods of 30 minutes. 1.05 x 10^{-3} is the constant propane emission rate along the 100 m hose (g m^{-1} s^{-1}). F_i (s m^{-2}) is the calculated dispersion factor and changes with time.

To calculate the emission factor (q) the total number of vehicles N_i (veh s^{-1}) circulating along the street is needed. The emission factor is equal to the total average emission Q_i divided by N_i. Therefore, the slope of a linear regression of the $F_i * N_i$ vs. C_i data is the traffic emission factor q (g veh^{-1} m^{-1} or mg veh^{-1} Km^{-1}). The intercept corresponds to the average background concentration (C_b). The correlation coefficient R from the linear regression is an estimation of the accuracy of the calculated q. F_i is independent of the pollutant type and it can be used to calculate the emission rates for any pollutant monitored.

4.2.5. Traffic counts

Traffic flow was continuously recorded with a video camera. The camera was placed at 16 m above street level in a private house (point 3 in figure 4.1), recorded videos were saved on a hard disk. Traffic volumes were counted manually after the measuring campaign. The fleet was divided into four vehicle categories for the counting: motorcycles, cars and vans (LDV: gasoline light duty vehicles); and busses and trucks (HDV: diesel heavy duty vehicles). Traffic counts were performed at intervals of 30 minutes from 10:00 to 22:00 for at least all the days when the tracer was released.

4.2.6. Weather information

A Wireless Vantage Pro2 Plus weather station (model 6163UK) was used to measure meteorological parameters. The weather station was installed during the campaign on the top of a 28-m high building, close to the measuring site (point 4 in figure 4.1). Meteorological parameters registered were: temperature, solar radiation, rain, atmospheric pressure, humidity, wind speed and wind direction.

4.3. RESULTS

4.3.1. Fleet information

About 6.5 million motorcycles, 330 thousand cars, 11 thousand trucks and 20 thousand buses were counted during the 30 days selected for the traffic volume assessment. Figure 4.3 shows the daily average variation of the fleet composition at the measurement site (%). The fleet is mostly made of light duty gasoline vehicles (motorcycles: 95%; passenger cars and vans: 4.5%). Only 0.5% of the fleet is made of heavy duty diesel vehicles. Moreover, this fleet composition follows a regular pattern along the day. Additional counting conducted at major roads in HCMC showed the same fleet distribution, indicating that the traffic composition at the measuring site is representative of the situation in the whole city (HOUTRANS, 2004). This distribution is rather typical for Vietnam and for many Asian cities where the main mode of transport is the motorcycle.
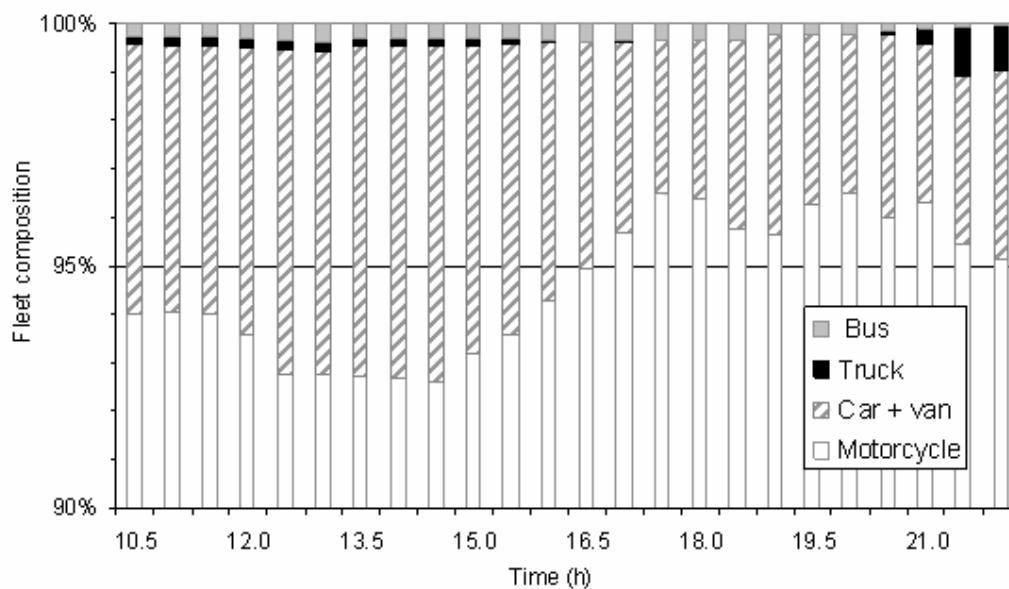

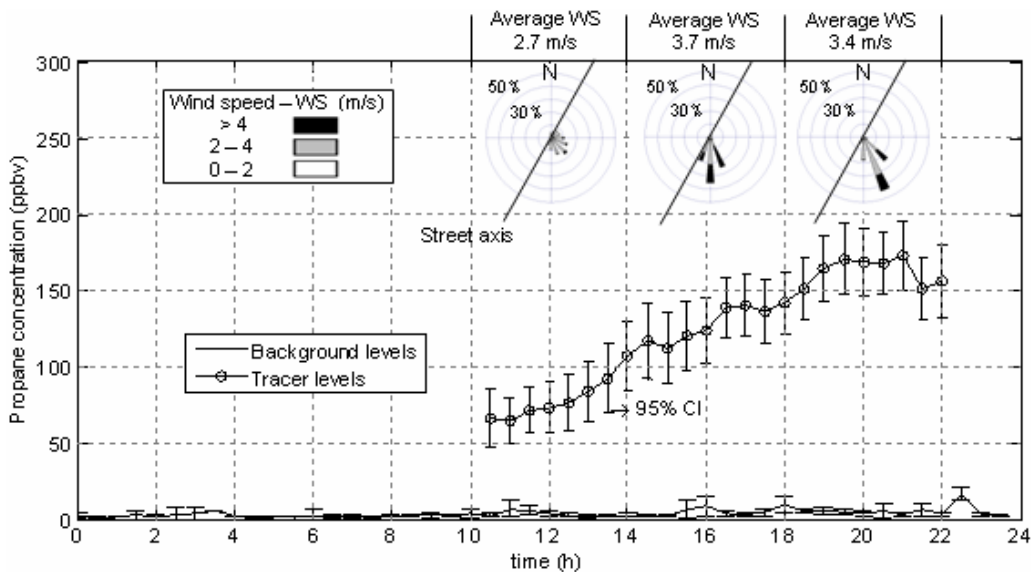Figure 4.3. Daily average variations of the fleet composition at the measurement site.

Figure 4.4. Average diurnal variations of propane based on all observed data without and with tracer release (95% CI: 95% confidence intervals). Wind roses at four-hour intervals (10:00-14:00; 14:00-18:00; 18:00-22:00) are shown at the upper part of the figure. The average wind speed (WS) for the four hours intervals is shown above the wind roses.

4.3.2. Tracer concentrations

LPG with the propane as tracer was released during about 300 hours on 25 non-consecutive days. At the same time 15 VOCs, NO, and $PM_{2.5}$ concentrations were monitored in 30 minutes intervals.

Figure 4.4 shows the average diurnal variations of propane based on all observed data without and with tracer release (identified respectively as background and tracer levels in the figure). The propane background levels are computed using every measurement (corresponding to a 30 minute interval) where there was no tracer released. This includes data from 35 days (of a total of 60 days of measurements) without any tracer release during the whole day and data from 25 days only for the hours without tracer release (22:30 to 9:30). The propane tracer levels are computed using every measurement where the tracer release was active. Figure 4.4 shows that the average propane backgrounds are typically below 10 ppbv. The average tracer levels during the release were well above the background propane levels present in the place (figure 4.4). For example, at 10:30, background propane levels are less than 5% of the values obtained during the tracer experiment. This percentage is even

smaller later in the day, which confirms that the effect of background propane concentrations on the tracer experiment can be neglected.

The tracer liberation was switched off every day exactly at 22:00. Thus, if there is an important amount of tracer remaining in the street after 22:00, this remaining propane should cause an increase in the calculated average background levels after this time. Therefore, the variation of the background propane levels after 22:00 gives an idea about the retention time of the tracer in the street. There are not important variations of background propane levels after this time indicating that the emitted tracer was rapidly transported outside the street (figure 4.4). Hence, there is no significant cumulative effect inside the street canyon. That is, an integrated 30-minute sample has no influence (or little) from the emission of the previous half an hour. A similar experimental study reported retention times in a street canyon to be below 4 minutes (De Paul and Sheih., 1985). However, results from measurement campaigns and model calculations have shown that in deeper street canyons the retention time can be as high as 4 hours (Murena and Ricciardi., 2005).

Figure 4.4 also shows wind roses drawn using wind speed and wind direction data registered at four-hour intervals. From 10:00 to 14:00, the wind blew from different directions and lower wind speeds conditions prevailed (the average wind speed is 2.7 m s^{-1}). The lowest tracer concentrations were observed at this period of the day. From 14:00 to 18:00, the wind direction was oblique to the street axis and wind speeds as well as tracer concentrations were higher than in the morning. From 18:00 to 22:00 the wind direction was almost perpendicular to the street axis and the average wind speeds was 3.4 m s^{-1}. Tracer concentrations from 18:00 to 22:00 were the highest observed.

The street canyon effect can be observed in figure 4.5. Different studies have shown that at high wind speeds and when the wind is perpendicular to the street axis, concentrations of pollutants increase at the leeward side of the street (Vardoulakis et al., 2003). The same behavior is observed in figure 4.5a. The increased leeward concentrations are due to the accumulation of pollutants locally advected by the wind vortex inside the street canyon. Tracer concentrations are higher at high wind speeds (figure 4.5b). Higher wind speeds occur when wind is perpendicular to the street (see figure 4.4) and thus concentrations are higher. This behavior has also been observed in other street canyon studies (Vardoulakis et al., 2007).
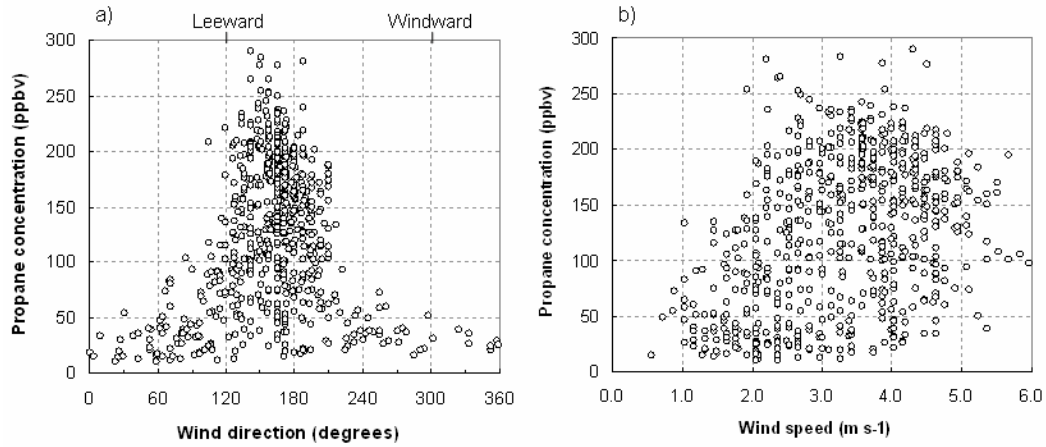
Figure 4.5. Propane (tracer) concentrations as a function of wind direction (a) and wind speed (b). The leeward and the windward indications in (a) denote the position of the measurement site in the street canyon with respect to the wind.
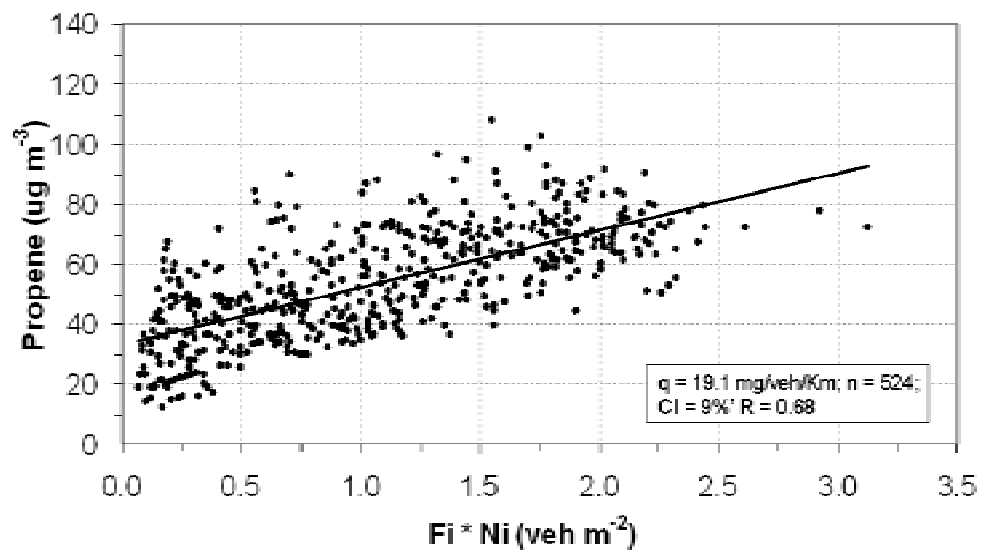

Figure 4.6. $F_i * N_i$ vs. propene concentrations and the estimated emission factor. The continuous line is the linear regression fit.

Table 4.1. Estimated traffic emission factors q (mg veh^{-1} Km^{-1}) and other parameters

Compound	n^a	R^b	q^c	CI^d %	C_b^e Ppbv	C^f ppbv	$(C - C_b) / C^g$ %
Propene	524	0.68	19.1 (6.7)	9	19.1	29.5	35
Trans-2-Butene	523	0.45	4.9 (1.7)	17	6.0	7.9	24
1-Butene	523	0.61	4.8 (1.7)	11	4.3	6.3	32
Cis-2-butene	521	0.44	4.6 (1.6)	17	5.7	7.5	24
i-Pentane	523	0.52	86.8 (30.4)	14	97.2	122.9	21
n-Pentane	522	0.63	27.0 (9.5)	11	25.8	33.7	23
Trans-2-Pentene	524	0.49	15.8 (5.5)	15	18.9	23.8	21
1-Pentene	522	0.58	5.5 (1.9)	12	4.3	5.9	27
2-methyl-2-butene	524	0.53	4.2 (1.5)	14	4.4	5.6	21
Cis-2-Pentene	524	0.56	5.3 (1.9)	12	4.0	5.6	29
2,3-Dimethylbutane	522	0.63	15.2 (5.3)	11	9.6	13.6	29
2-Methylpentane	523	0.58	14.6 (5.1)	12	9.2	12.8	28
3-Methylpentane	523	0.64	70.7 (24.7)	10	47.5	65.6	28
n-Hexane	524	0.47	116.9 (40.9)	16	106.2	136.5	22
Benzene	524	0.55	19.1 (6.7)	13	14.9	20.4	27
NO	401	0.31	39.3 (13.8)	30	101.5	135.6	25
$PM_{2.5}$ (ug/m3)	427	0.06	-	-	-	-	-

[a] Number of available data

[b] Correlation coefficient

[c] q: emission factors (values in parenthesis are the corrected emission factors, see section 4.3.4)

[d] 95 % confidence interval

[e] Average background concentration. Intercept form the C_i Vs. $F_i * N_i$ plot

[f] Total average pollutant concentration measured during the tracer experiment (10:00-22:00)

[g] Percentage of the pollutant directly emitted in the street $(C - C_b)$ to the total average pollutant concentration (C)

4.3.3. Traffic emission factors

We used the results of the tracer experiment to compute the dispersion factors (F_i) and then the traffic emission factors as explained in section 2.4. Figure 4.6 shows the F_i*N_i vs. the propene concentrations and its emission factor. Table 4.1 shows calculated emission factors for all the pollutants included in this study together with other important parameters. The emission factors were estimated from the slope of the F_i*N_i vs. C_i fitted with a linear regression model. The correlation coefficient R from the linear regression is an estimation of the accuracy of the calculated emission factors. Correlation coefficients presented in table 4.1 for the different pollutants are fair, nevertheless the emission factors are calculated from a large amount of data. The 95% confidence intervals (CI) are lower than CI found in other studies (Hwa et al., 2002; Kawashima et al., 2006; Chiang et al., 2007).

The method used in this study to estimate the emission factors is based in two assumptions: a) the emission factor q is constant with time and doesn't change due to e.g. changes in fleet composition and b) the background concentration $C_{b,i}$ is constant and doesn't show substantial daily variations. The fleet composition follows a regular pattern along the day (figure 4.3) and therefore the estimated emission factors are not affected by changes in vehicle fleet composition.

Ning et al. (2008) stated that emission factors estimated from roadside measurements are highly uncertain for species that are not characterized by substantial roadway emissions; it is, when their concentrations are similar both at freeway and at background sites. These background concentrations come from other sources than the traffic circulating on that specific street, i.e. other major roads nearby. Table 4.1 shows the background concentrations estimated from the intercept of the F_i*N_i vs. C_i best-fit-line. Table 4.1 also shows the total average pollutants concentration measured during the tracer experiment and the percentage of the pollutants directly emitted in the street with respect to the average total pollutant concentration. The contribution of the background is higher than the contribution of the emissions. Moreover, when the direct contribution is higher, the correlation coefficient tends to be higher as well (see table 4.1).

$PM_{2.5}$ and NO have the lowest correlation coefficients and then the highest uncertainties. The correlation coefficient for $PM_{2.5}$ is very low and the estimated emission factor has a negative slope. Thus the emission factor for this pollutant is not

reported. Hien et al. (2001) used a receptor model to identify the sources of $PM_{2.0}$ in HCMC. He found that only 17% of the $PM_{2.0}$ is directly emitted by road traffic. Therefore, $PM_{2.5}$ roadside levels should be very similar to background levels and thus, $PM_{2.5}$ emission factors can not be estimated.

In summary, to avoid the uncertainty produced by the background we recommend either to develop the measurements in a site with significant traffic emissions or either to measure the background concentrations.

4.3.4. Effect of the emission source position

The calculated F_i includes all the processes involved in the dispersion and transport of the tracer from the emission line to the measurement point. Pollutants emitted by traffic in the street are affected by the same dispersion processes and therefore, we assumed the calculated dispersion factors are also valid for all the pollutants emitted in the street. That is, we assumed that this emission line represents the emissions produced by traffic in the street. In reality, traffic emissions are produced over the full width of the street and then our assumption cause an error on the calculations.

Due to the absence of publications which have investigated the impact of such an error, we used the Computational Fluid dynamics model (CFD) WinMISKAM version 5.02 to estimate this error. Using this model, we calculated the dispersion factors (F_i) when an inert tracer substance is emitted from a line source placed on one side of the street. Estimations of the dispersion factors under the same conditions (meteorology, street geometry, etc.) but assuming the tracer is emitted from the whole street area (in the same way the traffic emissions are produced) were also developed.

The simulations show that the dispersion factors calculated for both source configurations are significantly affected by wind direction but they follow nearly the same trends. The dispersion coefficients calculated from the line source are in average 35% of those calculated from the area source, ranging from 30 to 40% depending on the wind direction. Here, as a first rough approximation, we used the average value of 35% as a correction factor for the emission factors. Table 4.1 shows the corrected emission factors. All the details about the CFD model set up and the estimation of this error will be presented in a separate paper.

Table 4.2. Comparison of corrected VOCs emission factors (mg veh^{-1} Km^{-1}) with other studies

Compound	HCMC [a] LDV: 99.5% HDV: 0.5% (MC: 95%)	Taipei [b] LDV: 90 - 96% HDV: 4 – 10%	Chung-Liao [c] LDV: 80 - 91% HDV: 9 – 20%	Taipei [d] MC
Propene	6.7	11.6	10.4	23
Trans-2-Butene	1.7	1.6	0.8	
1-Butene	1.7	8.3	10.7	
Cis-2-butene	1.6	1.8	1.6	
i-Pentane	30.4	12.5	40.1	118
n-Pentane	9.5	9.5	19.3	16
Trans-2-Pentene	5.5	2.8	4.1	
1-Pentene	1.9	1.6	1	
2-methyl-2-butene	1.5			
Cis-2-Pentene	1.9	1.6	1.6	
2,3-Dimethylbutane	5.3		12.7	
2-Methylpentane	5.1		12.6	22
3-Methylpentane	24.7		5.6	24
n-Hexane	40.9	4.18	5.7	
Benzene	6.7	12.2	5.9	20

LDV: Light Duty Vehicles (gasoline vehicles); HDV: Heavy Duty Vehicles (diesel vehicles); MC: Motorcycles (gasoline vehicles)

[a] This study

[b] Taipei tunnel study (Hwa et al., 2002)

[c] Chung-Liao tunnel (Chiang et al., 2007)

[d] Dynamometer test (Tsai et al., 2003). Emission factors extracted from figure 1 (in use 4-strokes motorcycles)

4.3.5. Comparison with available studies

Table 4.2 shows a comparison of corrected VOCs emission factors estimated here with other available studies. Since most of the vehicles in HCMC are light duty vehicles (LDV), results obtained here are compared to studies whit a high proportion of LDV in the fleet. Results are also compared to VOCs emission factors estimated for 4-stroke in use motorcycles. Most of the emission factors estimated in this study are within the range of emission factors reported in the other studies. The use of the CFD model to correct the EFs significantly improves the results but it is important to mention that the correction was made using an average value of 35% and that there

are also uncertainties associated to the model itself. These issues will be presented and discussed in a separate paper.

4.4. DISCUSSION AND CONCLUSIONS

In this study we designed and tested a system to estimate dilution factors and traffic emissions from a tracer study. We released continuously a passive tracer on one side of a busy street in Ho Chi Minh City, Vietnam. At the same time we measured on-line the resulting tracer concentrations as well as the concentrations of 15 VOC's, NO and $PM_{2.5}$ on the other side of the street.

We used propane contained in commercial LPG as a passive tracer; this choice resulted to be a good alternative to other common tracer substances even though there are some levels of this substance in the ambient air of the measurement site. Propane offers several advantages to traditional tracers: It is easily available, low price, non-reactive, negligible global warming potential, and easy to detect with commercial on-line gas chromatographs.

Tracer concentrations were smaller in the morning than in the afternoon and in the evening, it was observed that wind speed and wind direction played an important role in the tracer dispersion. This behavior was similar to results reported in other studies.

The results of the tracer experiment indicated that emitted tracer was rapidly transported outside of the street and that an integrated 30-minute tracer sample has no influence (or very little) from the emission produced during the previous half an hour. Thus, in this case the retention times were smaller than 30 minutes and they didn't influence the results of the tracer experiment. A similar set up can be used to experimentally estimate the retention times in other street canyons or even to validate existing dispersion models.

It was found that the pollutant background concentrations have an impact on the accuracy of the calculated emission factors. To avoid this impact, it is recommended either to develop the measurements at a site with significant traffic emissions or to measure the background concentrations.

We used the results of this tracer study to calculate the dilution factors and to estimate the traffic emission factors. The tracer was emitted on one side of the street from a finite emission line and measured at the other site of the street. We assumed that this emission line represents the emissions produced by traffic in the street. In reality, traffic emissions are produced over the area of the street lanes and then our assumption cause an error on the calculations. We used a Computational Fluid Dynamics model to calculate this error. Results show that the emission factors estimated from the area source are 35% of those calculate from the emission line.

The emission factors estimated in this study were compared with the emission factors estimated in other studies. The comparison shows that most of the VOCs emission factors estimated here are within the range of emission factors reported in other studies.

Up to our knowledge, this tracer study is one of the longest tracer experiments in a street canyon reported to date. The results from this work can be also used to validate existing dispersion models. This database is available to anyone interested.

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REFERENCES

Chiang, H., Hwu, C., Chen, S., Wu, M., Ma, S., Huang, Y., 2007. Emission factors and characteristics of criteria pollutants and volatile organic compounds (VOCs) in a freeway tunnel study. Science of the Total Environment 381, 200–211

Claiborn, C., Mitra, A., Adams, G., Bamesberger, L., Allwine, G., Kantamaneni, R., Lamb, B., Westberg, H., 1995. Evaluation of PM10 emission rates from paved and unpaved roads using tracer techniques. Atmospheric Environment 29, 1075-1089.

De Paul, F and Sheih, C., 1985. A tracer study of dispersion in an urban street canyon. Atmospheric Environment 19, 555-559

Gramotnev, G., Brown, R., Ristovski, Z., Hitchins, J., Morawska, L., 2002. Determination of average emission factors for vehicles on a busy road. Atmospheric Environment 37, 465-474.

Hien, P., Binh, N., Truong, Y., Ngo, N., Sieu, L., 2001. Comparative receptor modelling study of TSP, PM2.0 and PM2-10 in Ho Chi Minh City. Atmospheric Environment 35, 2669-2678.

Holmes, N., Morawska, L., 2006. A review of dispersion modeling and its application to the dispersion of particles: An overview of different dispersion models available. Atmospheric Environment 40, 5902–5928.

HOUTRNAS, 2004. The study on urban transport master plan and feasibility study in Ho Chi Minh metropolitan area. Volume 6, No 1 traffic and transport surveys. ALMEC Corporation.

Hueglin, C., Buchmann, B., Weber., R., 2006. Long-term observation of real-world road traffic emission factors on a motorway in Switzerland. Atmospheric Environment 40, 3696-3709.

Hwa, M., Hsieh, C., Wu, T., Chang, L., 2002. Real-world vehicle emissions and VOCs profile in the Taipei tunnel located at Taiwan Taipei area. Atmospheric Environment 36,1993–2002.

Kawashima, H., Minami, S., Hanai, Y., Fushimi, A., 2006. Volatile organic compound emission factors from roadside measurements. Atmospheric Environment 40, 2301-2312.

Ketzel, M., Wåhlin, P., Berkowicz, R., Palmgren, F., 2003. Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations. Atmospheric Environment, 37, 2735-2749.

Molina, M., Molina, L., 2004. Megacities and atmospheric pollution. Air and Waste Management Association 54, 644–680.

Murena, F and Ricciardi, G., 2005. CO residence times on urban roads in the Naples area using air quality monitoring data. Atmospheric Environment 39, 1993–2001.

Ning, Z., Polidori, A., Schauer, J., Sioutas, C., 2008. Emission factors of PM species based on freeway measurements and comparison with tunnel and dynamometer studies. Atmospheric Environment 42, 3099-3114.

Olcese, L., Palancar, G., Toselli, B., 2001. An inexpensive method to estimate CO and NOx emissions from mobile sources. Atmospheric Environment, 35, 6213-6218.

Palmgren, F., Berkowicz, R., Ziv, A., Hertel, O., 1999. Actual car fleet emissions estimated from urban air quality measurements and street pollution models. The Science of The Total Environment 235, 101-109.

Syntech., 2006. Manual for the Syntech Spectras GC955 series 400, 600, 800, single and double.

Tsai, J., Liu, Y., Yang, C., 2003. Volatile organic profiles and photochemical potentials from motorcycle engine exhaust. Air and Waste Management Association 53, 516-522.

Vardoulakis, S., Fisher, B., Pericleous, K., Gonzales, N., 2003. Modeling air quality in street canyons: a review. Atmospheric Environment 37, 155-182.

Vardoulakis, S., Valiantis, M., Milner, J., ApSimon, H., 2007. Operational air pollution modelling in the UK—Street canyon applications and challenges. Atmospheric Environment 41, 4622-4637.

Zárate, E., Belalcázar, Luis., Clappier, A., Manzi, V., Van den Bergh, H., 2007. Air quality modelling over Bogota, Colombia: Combined techniques to estimate and evaluate emission inventories. Atmospheric Environment 41, 6302-6318.

CHAPTER 5

IMPACT OF DIFFERENT SOURCE CONFIGURATIONS ON THE ESTIMATION OF VEHICLE EMISSION FACTORS FROM TRACER STUDIES

Abstract

In this work we use the Computational Fluid Dynamics Model (CFD) MISKAM to evaluate the impact of different source configurations on the estimation of road traffic emission factors (EFs) from tracer studies. This work is based on the results of a long term tracer experiment developed in Ho Chi Minh City (HCMC), Vietnam. In that study, a passive tracer was continuously emitted from a finite line source placed in one site of a street canyon. The results of the experiment were used in a previous work to calculate the dispersion factors and afterwards the EFs (Belalcazar et al., 2009). The proposed method assumes that the finite emission line represents the emission produced by traffic in the full area of the street and therefore there is an error associated to this assumption. Here we use the CFD model to calculate this error and to correct the HCMC EFs. In addition, the model is used to find a source configuration that better represents the vehicle emissions and that may be used in future studies to estimate more accurately the EFs. We also used the model to critically evaluate the proposed methodology.

First, the results of the tracer experiment are used to evaluate the performance of the CFD model on simulating the dispersion of the tracer. Results show that the model is able to simulate quite well the tracer dispersion in most of the cases. A comparison with available studies shows that the corrected HCMC EFs are within the range of the EFs reported in other studies. Different source configurations were also evaluated. Results show that a 200 m line placed in the center of the street would

represent very well the vehicle emissions. The simulations show that it is possible to accurately estimate the EFs from tracer studies.

Keywords: Computational Fluid Dynamics (CFD); tracer studies; real world emissions; traffic emission factors; street canyon

5.1. INTRODUCTION

In a previous work, the road traffic emission factors (EFs) were estimated from a long term tracer experiment carried out in Ho Chi Minh City (HCMC) Vietnam (Belalcazar et al., 2009). In the HCMC tracer experiment, a passive tracer substance was continuously emitted from a finite line source placed in one site of an urban street canyon. Simultaneously, the resulting tracer concentrations were monitored on-line at the other side of the street. The results of this experiment were used to calculate the dispersion factors and afterwards, these dispersion factors were used together with traffic counts and roadside pollutant measurements to estimate the EFs. This method assumes that the finite emission line represents the emission produced by traffic in the full width and length of the street and therefore there is an error associated to this assumption. In this paper, we use the Computation Fluid Dynamics Model (CFD) MISKAM to estimate this error and to correct the HCMC EFs.

In a first step we use the results of the tracer experiment to evaluate the performance of the CFD model on simulating the dispersion of the tracer in the street. After, the CFD model is used to calculate the dispersion factors when the tracer is emitted from the line source (case of the HCMC tracer experiment) and from the full length and width of the street, in the same way the traffic emissions are produced. The results of these simulations are used to compute the error produced by the source position and to correct the HCMC EFs. In addition, the model is also used to calculate the dispersion factors for other different hypothetical source configurations. The results of all these simulations are used to find the source configuration that better approximates to the road traffic emission and that may be used in future tracer studies to calculate more accurately the EFs.

5.2. METHODOLOGY

5.2.1. The Ho Chi Minh City (HCMC) tracer experiment

In this work we use the results of a long term tracer experiment which was conducted from January to March 2007 in the Ba Thang Hai street, close to the center of HCMC (Belalcazar et al., 2009). The tracer was emitted continuously from 10:00 to 22:00 during 25 non-consecutive days. n-propane was used as a passive tracer, it was emitted at a constant rate of 0.105 g s^{-1} from a 100 m perforated hose. The hose was placed at ground level between the west sidewalk and the lane at 8 m from the axis of the street (see 1 in figure 5.1a).

A mobile air quality monitoring stations placed at the other side of the street (receptor point) measured continuously the resulting tracer concentrations together with 15 Volatile Organic Compounds (VOCs) including the n-propane, NO, and $PM_{2.5}$ (point 2, figure 5.1a). Air samples were collected from the top of the monitoring station at a height of 2.5 m. Weather and traffic data were also continuously monitored during the experiment (points 3 and 4 in figure 5.1a)

5.2.2. Estimation of the EFs from a tracer study in HCMC

The results of the HCMC tracer experiment were used in a previous work to calculate the EFs (Belalcazar et al., 2009). Briefly, EFs were estimated by using the linear relation between pollutant concentrations and the vehicle emissions:

$$C_i = F_i \, N_i \, q + C_{b,i} \qquad\qquad (1)$$

Where C_i is the concentration of a particular pollutant in the street at any time i (µg m^{-3}). In this case, since the available pollutant concentrations are measured in 30-minutes intervals, i correspond to periods of 30 minutes. N_i is the total number of vehicles circulating along the street (veh s^{-1}); F_i is the dispersion factor (s m^{-2}) and $C_{b,i}$ is the background concentration (µg m^{-3}). q is the EF (g veh^{-1} m^{-1} or mg veh^{-1} Km^{-1}) calculated from the slope of the linear regression of the F_i * N_i vs. C_i plot. F_i was calculated from the tracer experiment as follows:

$$F_i = C_{t,\,i} \, / \, E \qquad\qquad (2)$$

Where $C_{t,i}$ is the concentration of tracer at the receptor point (μg m^{-3} or g (x 10^{-6}) m^{-3}) at the time i. E is the linear constant tracer emission rate, in the case of HCMC an emission rate of 1.05 x 10^{-3} g m^{-1} s^{-1} was used, it corresponds to the emission along the 100 m hose. Such estimation of F_i assumes that the finite emission line represents the emission produced by traffic in the full length and width of the street and that F_i doesn't depend on the source location and configuration. There is an error associated to this assumption. In order to evaluate the consecutive error, we use the CFD model to estimate this error and to correct the calculated EFs.
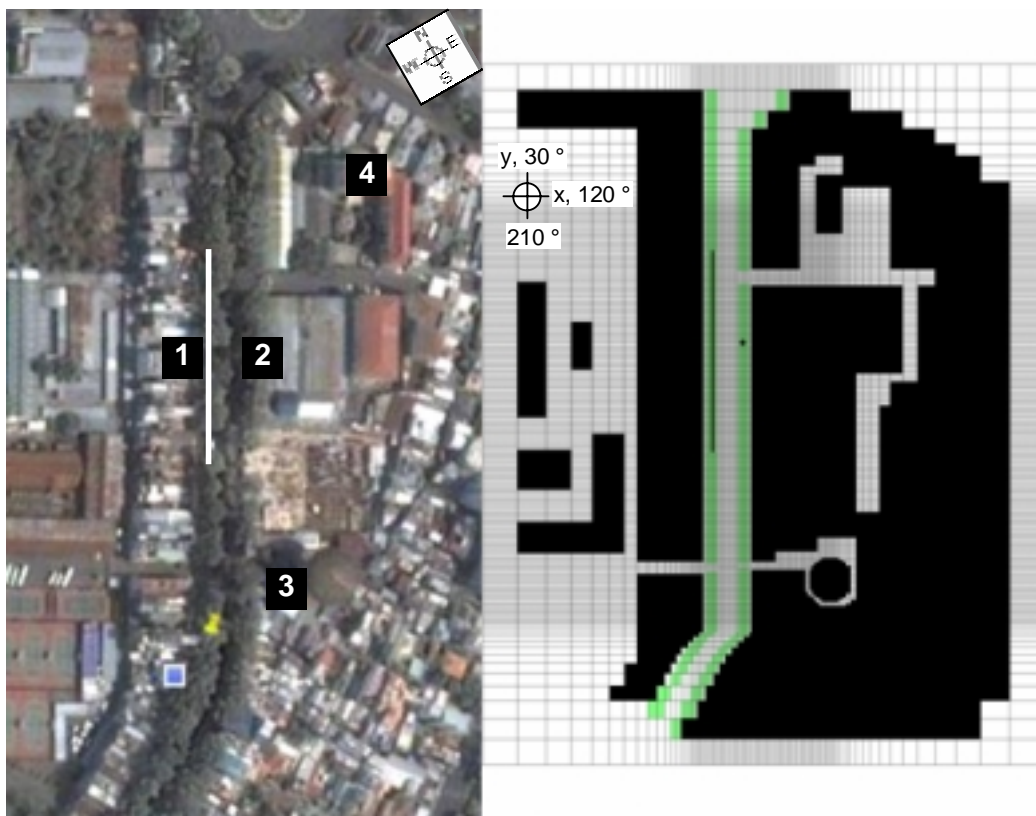
Figure 5.1. Left: (a) satellite Image of the Ba Thang Hai street. 1: emission line; 2: Monitoring station or receptor point; 3: Traffic video recording; 4: Meteorological station (source: Google Earth, downloaded in May 2009). Right: (b) WinMISKAM computational domain, the green cells indicate the presence of vegetation cover in that cell. The emission line and the receptor point are also indicated in the center of figure (b).

5.2.3. CFD model

WinMISKAM

WinMISKAM is the combination of user interfaces and the Model MISKAM (Microscale Climatic and Dispersion Model). It is a three-dimensional non-hydrostatic flow and dispersion model for microscale predictions of wind distribution and concentrations in urban areas (Eichhorn, J., 2008). The model solves the Reynolds Average Navier-Stokes (RANS) equations with the k-ε turbulent closure. This CFD model has been validated with wind tunnel data (Ketzel et al., 2000; Balczó et al., 2009; Olesen et al., 2009), and with field measurements collected in street canyons (Ketzel et al., 2000; Dixon et al., 2006).

MISKAM uses a staggered discretization grid of the type *Arakawa C*. Buildings are represented as rectangular block structures. The model first calculates the stationary wind fields in an Eulerian grid. After, the pollutants dispersion is calculated by means of an advection-diffusion approach. MISKAM includes a vegetation module which is able to consider the effect of vegetation on the flow field. This module has been recently evaluated with a dataset collected in a wind tunnel (Balczó et al., 2009).

On the other hand, as other CFD codes, MISKAM doesn't consider thermodynamic processes like energy transformation at the surface of the road, neither walls, roofs of buildings, nor thermal dispersion, buoyancy or water balance. This is mainly because it would significantly increase the computational time. The chemical transformation of pollutants is also not considered in the code. Moreover, a rough approach is used to consider the traffic induced turbulence. Although there are some experimental CFD models that include these parameters in their codes, further research is needed to correctly include and validate all these parameters in CFD models.

Computational domain

The domain selected for this work covers part of the Ba Thang Hai (BTH) street, it is close to the center of Ho Chi Minh City (HCMC), Vietnam. All the necessary information about this site was directly collected in the place or obtained from local governmental authorities. The BTH street is a highly transited two-way street canyon, it has three lanes each way. Their vehicle fleet is mostly made of light duty vehicles (motorcycles: 95%; cars and vans: 4.5%), 0.5% of the fleet are buses and trucks.

There are sidewalks at both sides of the BTH street of 4 meters width; the street including the sidewalks is 24 m width. The distance between the two closer intersections is about 350 m. There are 28 m trees standing at both sidewalks of the street (binomial name: Dipterocarpus Ablauts; family: Dipterocarpaceae). They are close one to the other and have crowns of 6 m average diameter at the last 8 m (see figure 5.1a and b).

The axis of the BTH Street is fixed to be near the center of the domain parallel to the y direction, it is aligned at 30° with respect to the north. A domain size of 280x350x60 m in the x, y and z direction was chosen for the simulations (figure 5.1b). Due to the long time needed for the simulations, this domain covers only 300 m out of the 350 m length of the street canyon. The impact of this domain size on the results will be discussed in section 5.3.1.

Buildings of 14 m average height stand beside the west sidewalks of the BTH street. Between these buildings and the left border of the domain there is a flat and open terrain partially used by some separated warehouses (figure 5.1). Five main structures stand out at the east and south-east of the BTH street. The first one is the Maximark supermarket (at the right of point 2 in figure 5.1a), it is a large rectangular building (52x73x14 m, x, y, z). There is also a 24 m water tower (at the right of point 3 in figure 5.1a), its mean diameter is 20 m. A military base in located in point 4, there is a building of 24 m height; the weather information was collected at the top of that building at 28 m height. Between the Maximark supermarket and the water tower there was another large supermarket of 52x53x12 m (x, y, z), but this construction was demolished in 2008, one year after the measuring campaign (see figure 5.1a). Buildings of 14–16 m height stand at both sides of the south and south-west sidewalks of the BTH street. Between points 2, 3 and 4 and the right border of the domain there are residential buildings of 6–8 m height.

Model set up

The domain is divided in 104x237x32 grid cells (x,y,z). Cells of different sizes are used in the x, y direction; the smallest size (1x1 m) is used along most of the surface of the BTH street. The dimensions of the cells progressively increase towards the borders of the domain (figure 5.1b). Five additional grid cells are added at the x,y borders of the domain with the same mesh width of the last cells (not shown in figure 5.1b). In the vertical direction (z) grid cells of 1 m height are used in the first 20 m of

the domain. After, the height of these cells increases gradually with a stretching factor of 1.18. This computational grid is specified in this way to better represent the buildings and the obstacles geometries present in the proximities of the BTH street.

All the simulations were carried out at neutral thermal stratification conditions (K/100m = 0). A ground roughness length of 0.1 m and a wall roughness length of 0.01 m are used for the simulations. These values have been proven to be suitable for CFD modeling in other studies (Dixon et al., 2006; Benson et al., 2008). The vegetation cover is also included in the computational domain (see figure 5.1b). A leaf area density of 0.45 m^2 m^{-3} is reported in the literature for trees of the Dipterocarpaceae family (Yoshimura et al., 2006), this value is used in the simulations.
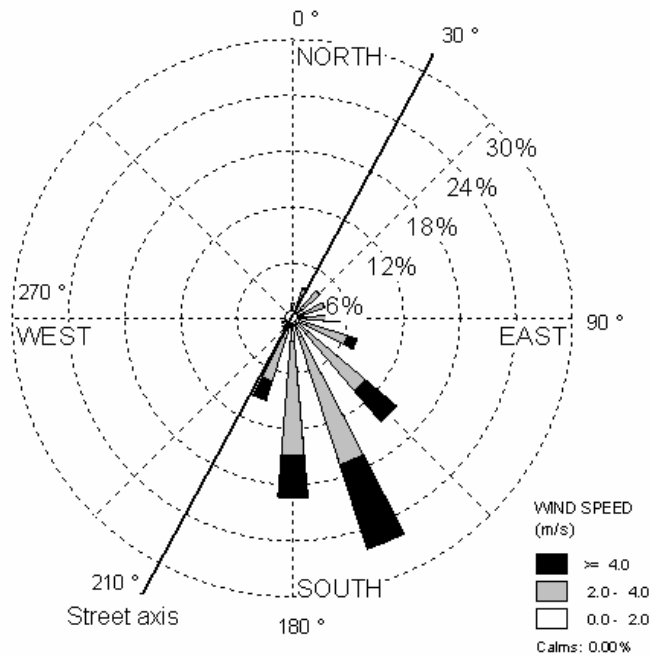
Figure 5.2. Wind rose plot for the HCMC tracer experiment (this plot was made from all the background wind directions and wind speeds observed at the measurement site during the tracer experiment)

MISKAM offers the possibility to compute the effect of the traffic induced turbulence. Different studies have shown that this parameter is important only at low wind speeds (Kastnerklein et al., 2003; Gidhagen et al., 2004; Dixon et al., 2006). Since in this case most of the background winds are above 2 m s^{-1} (figure 5.2) the effect of the

vehicle's turbulence shouldn't have an important effect on the results and therefore this parameter was not considered on the simulations.

CFD model performance

In order to simplify the evaluation process and to reduce the number of simulations, normalized concentrations are used to evaluate the performance of the model. All the tracer concentrations available are normalized as follow (Meroney et al., 1996):

$$C_i^* = C_i \, U_i \, H \, L \, / \, 0.105 \qquad\qquad (3)$$

Where C_i^* is the normalized concentration registered at the time *i*, since the tracer concentrations are measured in 30-minutes intervals, *i* correspond to periods of 30 minutes. C_i is the measured tracer concentration ($\mu g \ m^{-3}$ or $g(x10^{-6}) \ m^{-3}$), U_i is the measured background wind speed ($m \ s^{-1}$), H is the average height of the street canyon (14 m), L is the length of the canyon included in the domain (300 m), and 0.105 is the constant tracer emission rate ($g \ s^{-1}$). The normalized concentrations are grouped in wind direction clusters of 30°. The average normalized concentration and the standard deviation are calculated for each cluster.

As it is shown in figure 5.2, almost all the background winds come from 30 – 210 °, therefore, the model was run only for these wind directions at intervals of 30°. Since the background wind speed ranges from 2 to 6 m $^{-1}$, a wind speed of 3 m s^{-1} was used for the model evaluation. Modeled concentrations were also normalized using equation 3. Here, the tracer emission line and the receptor were set in the domain at the same position they were during the tracer experiment (see figure 5.1b).

5.2.4. Evaluation of emission source configurations

Evaluation of the source configuration used in HCMC

Here we use the CFD model to evaluate the effect of the source configuration used in HCMC on the estimated EFs and to estimate the error produce. First, the model is used to estimate the tracer concentrations at the receptor point for the real case of HCMC, it is, emission line placed beside the left sidewalk at ground level. After, and in order to represent the vehicle emissions, the model is also used to calculate the tracer concentrations at the receptor point by using the same conditions
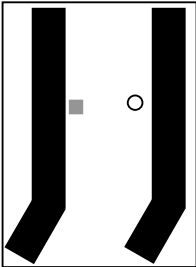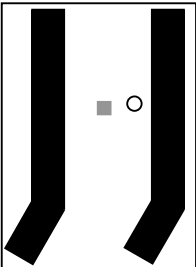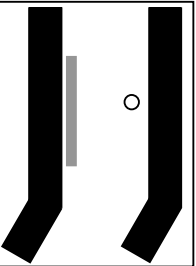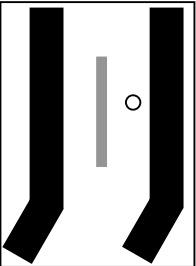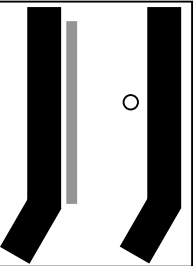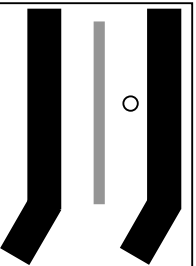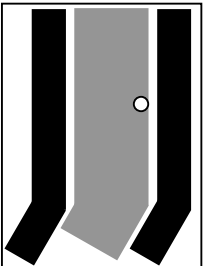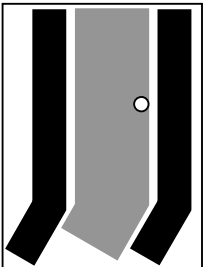
(meteorology, street geometry, etc.) but assuming the tracer is emitted at ground level from the full street area (full street length and width but excluding the sidewalks). Kumar et al., (2009) found that the emission source height (in the z direction) has an impact on the modeled concentrations at the lower part of the street canyon. Therefore, we also use a volume source with a height of 2 m (z= 0 – 2 m). Table 5.1 summarizes the details about these source configurations. In the case of the area and the volume sources, these emissions are multiplied by the width and the height (z) of the emission source to get the emission in g m^{-1} s^{-1}. In all the cases the same total tracer emission is used (0.105 g s^{-1}).

Next, the dispersion factors for each source configurations are calculated by using equation (2). All the simulations are developed at wind direction intervals of 30°. Since the predominant wind speed ranges between 2–6 m s^{-1} (see figure 5.2), two sets of simulations were carried out for the different wind direction intervals, one at 3 and the other one at 5 m s^{-1}. The difference between the dispersion factors estimated for the source used in HCMC and the dispersion factors estimated for the source that represents the traffic emissions (area or volume) corresponds to the error produced by the source.

Evaluation of other source configurations

Here other hypothetical source configurations are evaluated (table 5.1). These sources may be used in future studies to estimate more accurately the EFs. Since these emission sources evaluated have different lengths, the linear emission (E) changes depending on the source configuration but in all the cases also the same total tracer emission is used (0.105 g s^{-1}). Here the tracer concentrations are estimated at the receptor point for each source configuration and the dispersion factors are calculated from equation (2). These simulations are also developed at wind direction intervals of 30° and at 3 and 5 m s^{-1}.

Table 5.1. Emission source configurations evaluated

Source	Source position in the street [a]		Width, length,	Emission
	Left	Center	x,y (m)	Rate (E)
Point			1,1	0.105 $g\ m^{-1}\ s^{-1}$
100 m line	[b]		1,100	1.05×10^{-3} $g\ m^{-1}\ s^{-1}$
200 m line			1,200	5.25×10^{-4} $g\ m^{-1}\ s^{-1}$
Area	[c]		16,300	2.19×10^{-5} $g\ m^{-2}\ s^{-1}$ or 3.5×10^{-4} $g\ m\ s^{-1}$
Volume (z = 0-2 m)	[c]		16,300	1.09×10^{-5} $g\ m^{-3}\ s^{-1}$ or 3.5×10^{-4} $g\ m\ s^{-1}$

[a] Black: buildings; Grey: source position; White circle: receptor position

[b] This configuration corresponds to the HCMC tracer experiment (real case)

[c] Sources that represent the vehicle emissions

5.2.5. Correction of the HCMC EFs

The difference between the dispersion factors calculated from the base case and from the sources that represent the vehicle emissions corresponds to the error produced by the source position and it is used to correct the dispersion factors used in HCMC to estimate the EFs. The corrected dispersion factors are then used to recalculate the EFs by using equation (1) and the procedure described at the beginning of the previous section.

5.3. RESULTS

5.3.1. CFD model performance

Figure 5.3 shows the modeled and the observed normalized concentrations as a function of wind directions. As can be seen, MISKAM is able to reproduce quite well the results of the HCMC tracer experiment. The modeled values follow the same average trend of the observations. The highest observed values are from 120 to 180°, been the highest at 150°, MISKAM also reproduces this behavior and also predicts the highest concentration at 150°.

There is an outstanding performance of the model when wind is oblique or perpendicular to the street axis (60 - 180°). The modeled concentration at 120° match with the average observed values (figure 5.3). At 60, 90 and 150° the modeled normalized concentrations are within the standard deviations of the observations but they are slightly underestimated, in average they are 24% smaller than the observed values. Figure 5.4 shows that when wind blows from 120° it enters into the street and hits the west buildings, then it changes its direction and carries the tracer to the receptor point. A similar behavior was observed at 60, 90, 150 and 180°.

On the other hand, the model underestimates the normalized concentrations when wind is parallel to the street axis; in particular it is significantly underestimated at 210°. At this direction wind flows parallel to the street and pushes the tracer downstream, without reaching the receptor point (figure 5.4). As it was previously stated in section 5.2.2, the selected domain only covers 300 m of the BTH street, it only covers part of the buildings that are at the south-west of the domain (figure 5.1).
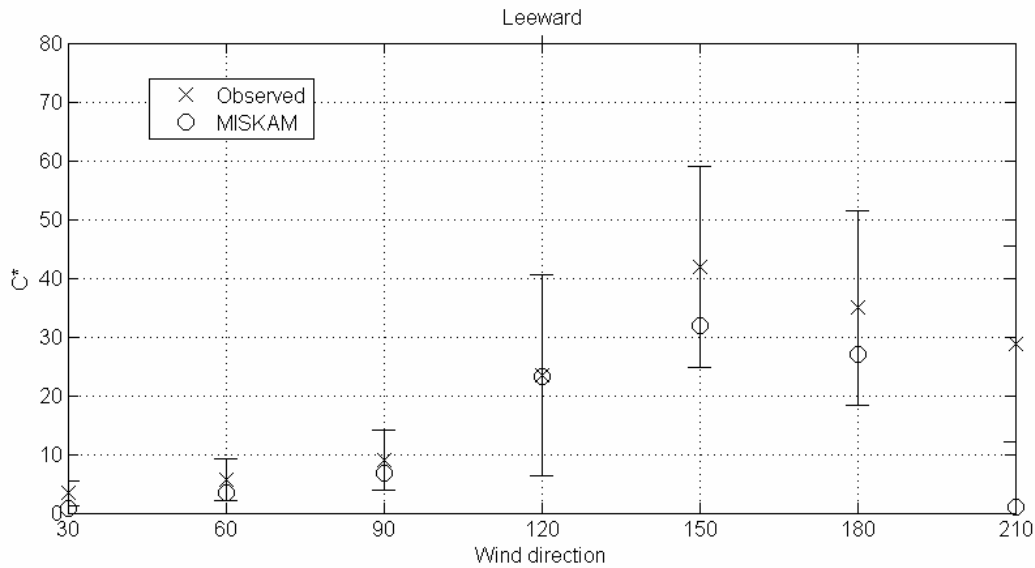
Figure 5.3. Modeled and observed normalized concentrations of tracer as a function of background wind direction. X corresponds to the average observed normalized values, they were calculated from all the data available at the indicated wind direction (±15°). The standard deviations of the observed values are shown as vertical bars.

These buildings shelter the street and change the flow direction which carries the tracer to the receptor (as it happens at 180°, see figure 5.4). A larger domain would improve the results at 210° but it would also increase the computation time. Anyhow, in this case most of the data were registered for wind directions between 60-180° (see figure 5.2); therefore the subsequent simulations are carried out only at this wind directions.

As it can be seen, MISKAM was able to reproduce very well the average behavior of the tracer in the street canyon even though it doesn't consider thermodynamic processes such as energy transformation at the surface of the road, neither walls, roofs of buildings, nor thermal dispersion, buoyancy or water balance. In the case of HCMC, the CFD model produced very good results without considering this parameter on the simulations. It will be necessary to develop similar studies in other cities of the world and to quantify the real impact of this parameter on the simulation results.
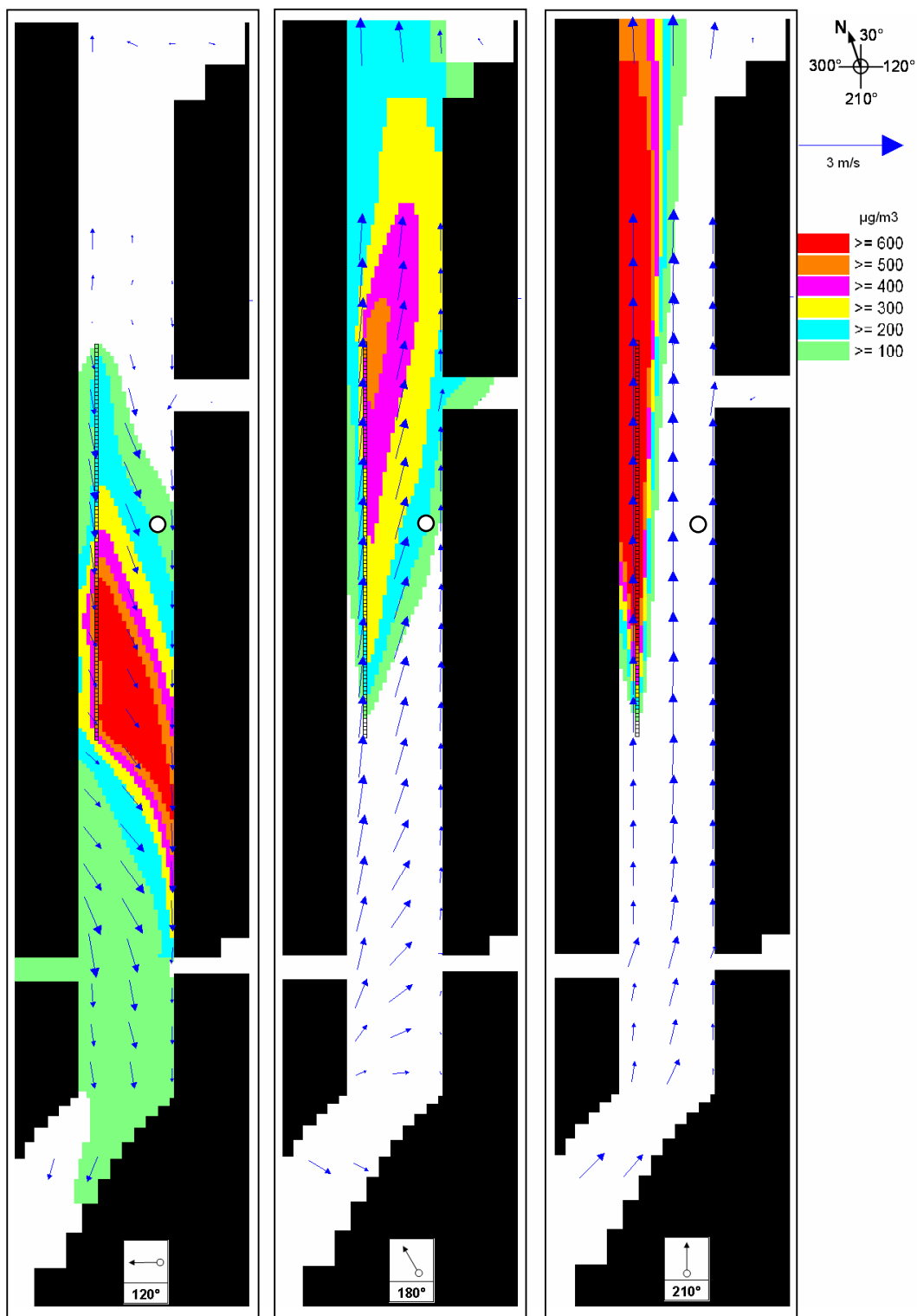
Figure 5.4. Tracer distributions over the BTH street at the receptor level (z = 2–3 m). The black arrows at the bottom of each figure indicate the background wind direction used for the simulation. The emission line and the receptor position are indicated in the center of the figures.

It is important to mention here that most of the research conducted to date used theoretical building configurations or measurements collected in reduced scale laboratory experiments to evaluate the models (Li et al., 2006; Benson et al., 2008). In this work we use a long term tracer experiment which was developed in real urban conditions to evaluate the CFD model. Moreover, an important limitation of CFD model evaluation in real urban conditions has been the uncertainties in the input information such as the vehicle emission factors (EFs) (Lohmeyer et al., 2002; Vardoulakis et al., 2003; Gidhagen et al., 2004; Dixon et al., 2006; Holmes et al., 2006). The lack of accurate EFs has difficult the validation of dispersion models. Here the uncertainty associated to the emissions was eliminated from the evaluation process because the tracer emission rate is known.

5.3.2. Evaluation of different source configurations

Eight source configurations are evaluated; figure 5.5 shows the dispersion factors (F) calculated from equation (2) for all these sources. Two of the sources (area and volume) are used to represent the vehicle emissions. The dispersion factors calculated from these sources are practically the same for almost all wind directions and speeds (figure 5.5). Only at 60° and at 3 m s^{-1}, F for the volume source is 7% smaller than the area source but in the rest of the cases this difference is smaller than 5%. Therefore, in this case any of these sources is able to represent the vehicle emissions. In order to simplify the explanations from now to ahead only the volume source is used.

Wind speed is inversely related to F. At higher wind speed the tracer concentrations at the receptor are smaller and then F is also smaller, this behavior is observed for all the source configurations and for all wind directions (figure 5.5). F strongly depends on wind direction. All the F calculated from most of the source configurations used are smaller than the F calculated from the volume source. The smallest differences between all the evaluated sources and the volume source occur when wind is perpendicular or almost perpendicular to the street axis (90-150°). The largest differences occur when wind is oblique to the street axis (60 and 180°), at these wind directions the difference is larger for the sources that are at the left of the street.
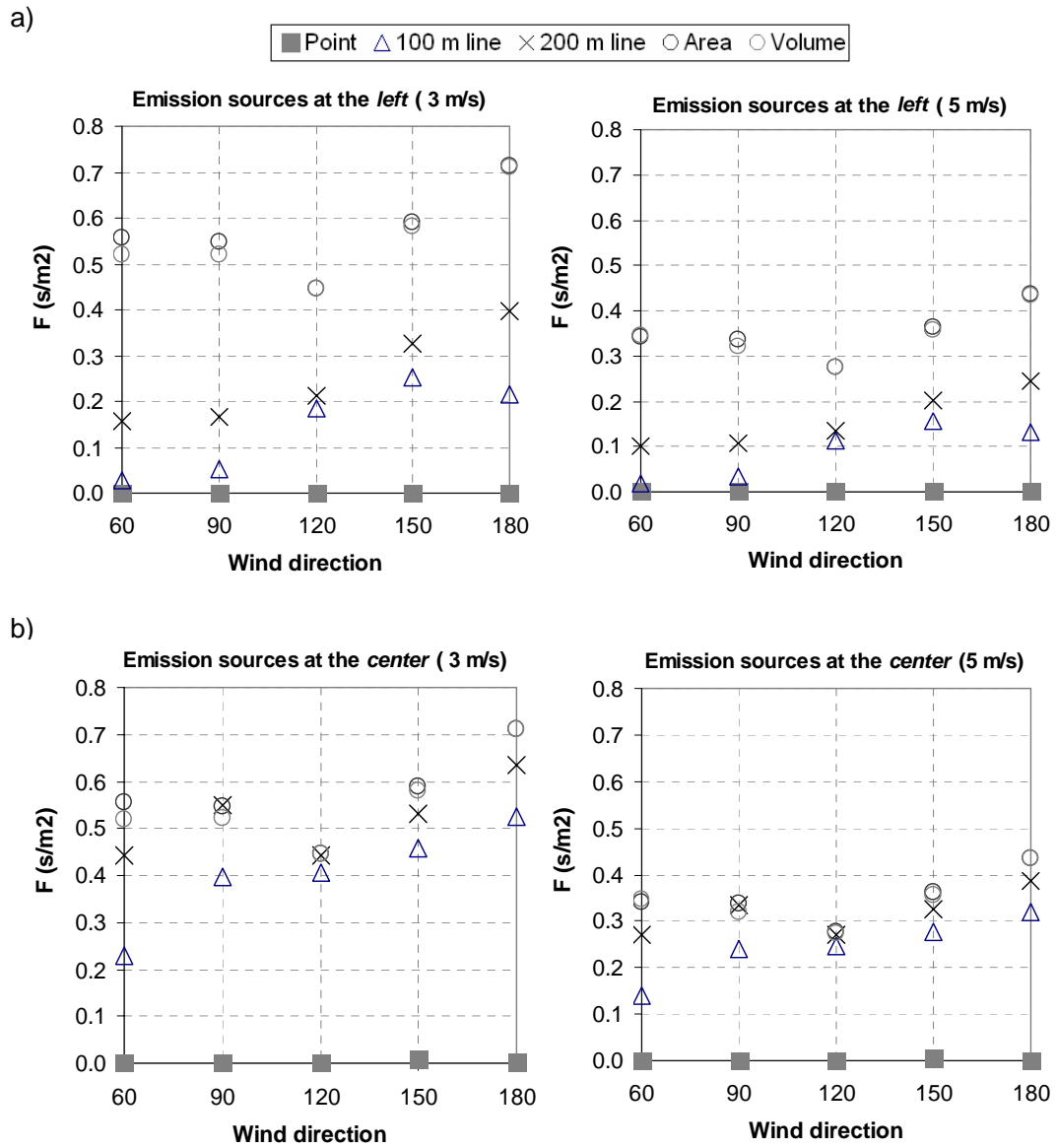
a)

b)

Figure 5.5. Dispersion factors (F, s m^{-2}) as a function of wind directions. a) Emission sources at the left of the street (the *100 m line* corresponds to the real case of HCMC). b) Emission sources at the center of the street (hypothetical emission sources). In both cases the model was run at 3 and at 5 m s^{-1}.

Figure 5.5 shows that the point source is the configuration that less approximates to the vehicle emissions. F calculated from this source are more than 95% below the F calculated from the volume source in all the cases (Figure 5.5a 5.5b). Figure 5.5a shows F calculated from the HCMC tracer experiment. For some wind directions F calculated from this source nearly follows the same trends of the volume source. From 120-180°, the difference between the F calculated from these sources is 56-70% (see table 5.2) whereas at 60-90° it is 89-95%. A 200 m line source placed at the left of the street would improve the results at 60 and 180° but at the other wind directions the error would keep nearly the same.

The sources that better approximate to the vehicle emissions are the ones that are placed in the center of the street. A 100 m line source placed there would give better results than even a 200 m line placed in the left of the street. The source that represents the best the vehicle emissions is the 200 m line placed in the center of the street (see figure 5.5 and table 5.2). For almost all the wind directions the difference between the F calculated from this source and the volume source is smaller than 10%. Only at 60° and 5 m s^{-1} this difference is 21%.

Table 5.2. Difference (error, %) between the sources configurations evaluated and the volume source (the source that represents the vehicle emissions)

Wind direction	100 m line at the *left* of the street [b]		200 m line at the *left* of the street		100 m line at the *center* of the street		200 m line at the *center* of the street	
	3 m s^{-1}	5 m s^{-1}	3 m s^{-1}	5 m s^{-1}	3 m s^{-1}	5 m s^{-1}	3 m s^{-1}	5 m s^{-1}
60	95	95	70	71	56	60	14	21
90	90	89	68	67	24	25	- 5	- 4
120	59	59	52	51	9	11	0	2
150	56	57	44	44	21	22	8	9
180	70	70	44	44	26	27	11	11

[a] The difference is calculated as: $(F_{volume} - F_{source}) / F_{volume}$

[b] Real case in the HCMC tracer experiment

5.3.3. Correction of the HCMC EFs

The results presented in the previous section are used to correct the EFs factors calculated from the HCMC tracer experiment. As can be seen in table 5.2, the errors calculated for this source are independent of wind speed and therefore the correction can be applied to any wind speed. However, the differences strongly depend on wind direction. The dispersion factors are corrected by using the next expression:

$$F_{i,c} = F_i / (1 - error/100) \tag{4}$$

F_i is the dispersion factor that was initially calculated from the tracer experiment (s m^{-2}). The error is taken from table 5.2 for the respective wind direction. Since the smallest errors are presented at 120 and 150° (table 5.2), only the F_i belonging to this directions (±15°) are used. $F_{i,c}$ is the corrected dispersion factor. The EFs are then recalculated from equation (1) and the procedure described in section 5.2.2.
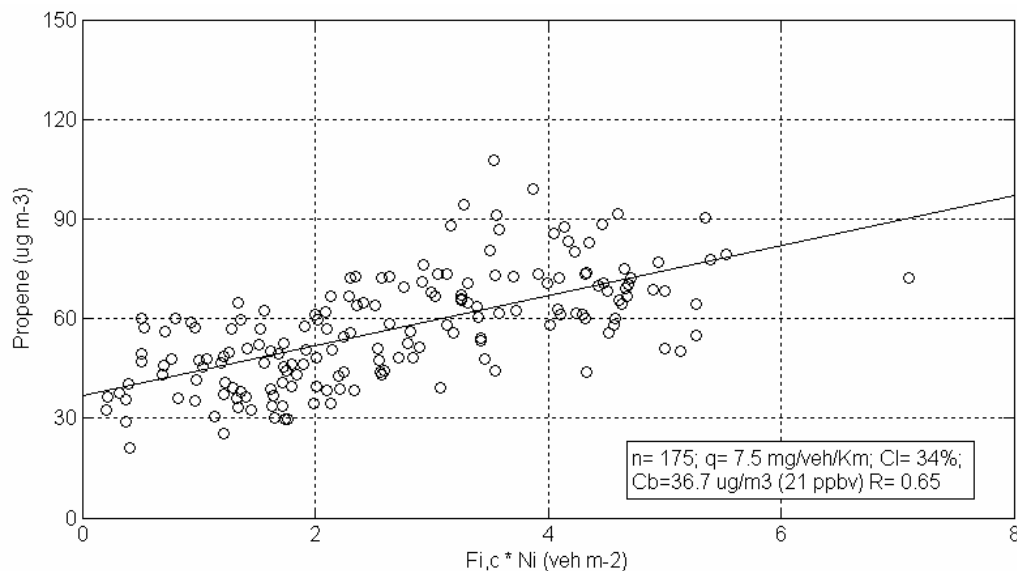

Figure 5.6. Corrected propene vehicle emission factors (the continuous line is the linear regression fit. CI is the 95% confidence interval).

Table 5.3. HCMC VOCs emission factors (q, mg veh^{-1} Km^{-1}) and comparison with other studies

Compound	HCMC [a]				Taipei[b]	Chung-Liao[c]	Taipeic[d]
	n	C_b	R	q (CI)			
Propene	175	21.0	0.65	7.5 (35%)	11.6	10.4	23
Trans-2-Butene	175	6.1	0.47	2.0 (57%)	1.6	0.8	
1-Butene	175	4.8	0.58	1.8 (42%)	8.3	10.7	
Cis-2-butene	174	5.7	0.47	1.9 (57%)	1.8	1.6	
i-Pentane	175	102.2	0.51	33.9 (51%)	12.5	40.1	118
n-Pentane	175	28.8	0.57	9.7 (43%)	9.5	19.3	16
Trans-2-Pentene	175	18.9	0.52	7.4 (49%)	2.8	4.1	
1-Pentene	173	4.6	0.57	2.3 (43%)	1.6	1	
2-methyl-2-butene	175	4.6	0.57	1.9 (44%)			
Cis-2-Pentene	175	4.4	0.55	2.1 (45%)	1.6	1.6	
2,3-Dimethylbutane	174	11.1	0.56	5.2 (44%)		12.7	
2-Methylpentane	174	10.5	0.44	5.6 (61%)		12.6	22
3-Methylpentane	175	51.9	0.62	27.2 (38%)		5.6	24
n-Hexane	175	105.5	0.51	55.9 (51%)	4.18	5.7	
Benzene	175	16.5	0.51	7.5 (51%)	12.2	5.9	20

LDV: Light Duty Vehicles (gasoline vehicles); HDV: Heavy Duty Vehicles (diesel vehicles); MC: Motorcycles (gasoline vehicles)

[a] This study. n: Available number of data at the selected wind directions (105-165°); C_b: Background concentration (ppbv) R: correlation coefficient; CI: 95% confidence intervals. LDV: 99.5% (MC: 95%); HDV: 0.5%

[b] Taipei tunnel study (Hwa et al., 2002). LDV: 93%; HDV: 7%

[c] Chung-Liao tunnel (Chiang et al., 2007). LDV: 85%; HDV: 15%

[d] Dynamometer test (Tsai et al., 2003). Emission factors extracted from figure 1 (in use 4-strokes motorcycles)

Figure 5.6 and table 5.3 present the corrected EFs together with some statistical parameters. The correlation coefficient (R) and the background concentration (C_b) keep nearly the same to the ones calculated before (see Belalcazar et al., 2009). Conversely, the confidence intervals are higher. Note that here only the data from selected wind directions are used (105-165°), the EFs are calculated only from 33% of the total available data. This reduction may explain the increase on the confidence interval. It is important to mention that the confidence intervals of the EFs are typically not reported. In this work it is possible to do it because a large dataset was collected with the on-line gas chromatograph.

Table 5.3 also presents a comparison of the corrected VOCs EFs with other studies. As can be seen almost all the EFs estimated in this study are within the range of the EFs reported in other studies. Only the hexane EF is well above the EFs reported in the other studies.

5.4. CONCLUSIONS

In this work we use the Computational Fluid Dynamics Model (CFD) MISKAM to evaluate the impact of different source configurations on the estimation of road traffic emission factors (EFs) from tracer studies. We used the CFD model to calculate the error produced by the source configurations on the dispersion factors and therefore on the estimated EFs. In addition, we also used the model to find a tracer source configuration that better represents the vehicle emissions.

In a first step, the results of the tracer experiment were used to evaluate the performance of the CFD model on simulating the dispersion of the tracer. The results of this evaluation indicate that the model is able to simulate quite well the tracer dispersion in this urban environment. The model reproduces the same average trends and levels of the observations in most of the cases. There is a very good performance when wind is perpendicular or oblique to the street axis but the model underestimates the concentrations when wind is parallel to the street axis. This underestimation is attributed to the size of the chosen simulation domain.

The model was also used to calculate the error produced by the source configuration used in HCMC to estimate the EFs. The calculations show that the error is smaller when wind is perpendicular or almost perpendicular to the street. These errors were afterwards used to correct the dispersion factors and to recalculate the HCMC EFs. A comparison with available studies indicates that the corrected emission factors are within the range of the EFs reported in other studies.

In order to find the source configuration that better represents the vehicle emissions, the CFD model was run using the same conditions (meteorology, street geometry) but changing the source configurations. Three source configurations placed in two different positions are evaluated; they are a point source, a 100 m line source and a 200 m line source. All these configurations were placed in one side and in the center

of the street. These configurations included the one used in the HCMC tracer experiment (100 m line placed in one side of the street). The simulation results show that a 200 m line place in the center of the street would represent very well the vehicle emissions. It would be very interesting to experimentally test this source configuration and to use it to estimate the EFs in a city.

REFERENCES

Balczó, M., Gromke, Ch., Ruck, B., 2009. Numerical modeling of flow and pollutant dispersion in street canyons with tree planting, Meteorologische Zeitschrift, Vol. 18, 197 – 206.

Belalcazar, L., Fuhrer, O., Ho. D., Zarate, E., Clappier, A., 2009. Estimation of road traffic emission factors from a long term tracer study in Ho Chi Minh City (Vietnam). Atmospheric Environment. In Press, Accepted Manuscript.

Benson, J., Ziehn, T., Dixon, N.S., Tomlin, A.S., 2008. Global sensitivity analysis of a 3-dimensional street canyon model-Part II: Application and physical insight using sensitivity analysis. Atmospheric Environment 42, 1874-1891.

Chiang, H., Hwu, C., Chen, S., Wu, M., Ma, S., Huang, Y., 2007. Emission factors and characteristics of criteria pollutants and volatile organic compounds (VOCs) in a freeway tunnel study. Science of the Total Environment 381, 200–211

Dixon, N.S., Boddy, J.W.D., Smalley, R.J., Tomlin, A.S., 2006. Evaluation of a turbulent flow and dispersion model in a typical street canyon in York, UK. Atmospheric Environment 40, 958-972.

Eichhorn, J., 2008. WinMISKAM for Windows manual (version 5.02). Institute for Atmospheric Physics, University of Mainz, Germany

Gidhagen, L., Johansson, C., Langner, J., Olivares, G., 2004. imulation of NOx and ultrafine particles in a street canyon in Stockholm, Sweden. Atmospheric Environment 38, 2029-2044.

Holmes, N., Morawska, L., 2006. A review of dispersion modeling and its application to the dispersion of particles: An overview of different dispersion models available. Atmospheric Environment 40, 5902–5928.

Hwa, M., Hsieh, C., Wu, T., Chang, L., 2002. Real-world vehicle emissions and VOCs profile in the Taipei tunnel located at Taiwan Taipei area. Atmospheric Environment 36,1993–2002.

Kastner-Klein, P., Fedorovich, E., Ketzel, M., Berkowicz, R., Britter, R., 2003. The modelling of turbulence from traffic in urban dispersion models - Part II: Evaluation against laboratory and full-scale concentration measurements in street canyons. Environmental Fluid Mechanics 3, 145-172.

Ketzel, M., Berkowicz, R., Lohmeyer, A., 2000. Comparison of numerical street dispersion models with results from wind tunnel and field measurements. Environmental Monitoring and Assessment 65, 363-370.

Kumar, P., Garmory, A., Ketzel, M., Berkowicz, R., Britter, R., 2009. Comparative study of measured and modelled number concentrations of nanoparticles in an urban street canyon. Atmospheric Environment 43, 949-958.

Li, X., Liu, C., Leung, D., Lam, K., 2006. Recent progress in CFD modelling of wind field and pollutant transport in street canyons. Atmospheric Environment 40, 5640–5658

Lohmeyer, A., Mueller, W.J., Baechlin, W., 2002. A comparison of street canyon concentration predictions by different modelers: Final results now available from the Podbi-exercise. Atmospheric Environment 36, 157-158.

Meroney, R.N., Pavageau, M., Rafailidis, S., Schatzmann, M., 1996. Study of line source characteristics for 2-D physical modelling of pollutant dispersion in street canyons. Journal of Wind Engineering and Industrial Aerodynamics 62, 37-56.

Olesen, H.R., Berkowicz, R., Ketzel, M., Løfstrøm, P., 2009. Validation of OML, AERMOD/PRIME and MISKAM using the Thompson wind-tunnel dataset for simple stack-building configurations. Boundary-Layer Meteorology 131, 73-83.

Tsai, J., Liu, Y., Yang, C., 2003. Volatile organic profiles and photochemical potentials from motorcycle engine exhaust. Air and Waste Management Association 53, 516-522.

Vardoulakis, S., Fisher, B.E.A., Pericleous, K., Gonzalez-Flesca, N., 2003. Modelling air quality in street canyons: A review. Atmospheric Environment 37, 155–182

Yoshimura, M., Yamashita, M., Ichie, t., 2006. Light environmental monitoring in tropical rainforest. Proceedings of the Asian Conference on Remote Sensing (ACRC).

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE RESEARCH WORKS

6.1. CONCLUSIONS

Road traffic emissions are highly uncertain in many cities of the developing world. Existing techniques to assess vehicle emissions are expensive and not always accurate, therefore adequate abatement strategies can not be adopted in such cities. In this PhD thesis alternative techniques to assess road traffic emissions were developed. The techniques were implemented in Ho Chi Minh City (HCMC), Vietnam. Due to the developing conditions of the city and especially to the lack of the necessary funding to correctly assess its traffic emissions, this city resulted in an interesting place to test these techniques. This work is based on an intensive measuring campaign that took place in an urban street canyon located in the center of HCMC. For a first time in this city, a state-of-the-art on-line gas chromatograph was used to measure the roadside levels of 19 C_2 – C_6 Volatile Organic Compounds (VOCs). Other cutting edge devices were also used to measure on-line the NO and $PM_{2.5}$ concentrations.

In a first stage, the results of the measurements were used to assess the roadside levels of VOCs and the other pollutants in HCMC and to identify the sources of these pollutants. This part of the research let to extract the next conclusions:

- There is a severe air pollution problem in HCMC. A comparison with available studies demonstrates that selected VOCs levels are the highest roadside levels reported in Asia. Moreover, pollutant concentrations are well above the available World Health Organization Guidelines (WHO).

- From the VOCs monitored, the most abundant species are n-hexane, i-pentane, and 3-methylpentane; they represent 60% of the total VOCs monitored.

- The Maximum Incremental Reactivity (MIR) was used to compute the Ozone Formation Potential (OFP) of the measured VOCs. Trans-2-pentene, i-pentane, propene and n-hexane have the highest OFP, these four VOCs have the potential to generate 44% of the total ozone produced by all the VOCs included in this study.

- A Principal Components Analysis was used to identify the sources of the pollutants. Results show that all the measured VOCs (except isoprene) are produced by gasoline powered vehicles (motorcycles), whereas NO is associated to diesel powered vehicles (buses and trucks). This analysis also reveals that road traffic is not an important direct source of $PM_{2.5}$ in HCMC. Additional research is required to identify the sources of particles in the city.

- Despite motorcycles offer a number of advantages in terms of transportation, this study shows that this source is responsible of the elevated levels of VOCs. Authorities should develop other transportation modes and encourage their use.

- The use of the on-line roadside pollutants monitoring together with the Principal Components Analysis showed to be a good first approach to assess the road traffic emission. This is an affordable approach and it is strongly recommended for preliminary studies in cities with limited financial means to tackle the air pollution problem.

After, an innovative technique to estimate the road traffic emission factors (EFs) was developed and tested. The EFs were estimated from a long term tracer experiment. A continuous tracer emission system was developed and implemented. The system consists of two parts: 1) A finite line source placed in one side of the urban street canyon where the tracer is continuously emitted at a constant and known rate. 2) A gas chromatograph which is placed at the other site of the street and which measures on-line the resulting tracer concentrations. This system was tested in the HCMC campaign. The conclusions from this part of the research are:

- The tests of the tracer emission system show that it is possible to reproduce the vehicle emissions with such a system. In addition, there are many potential applications of this device in the urban air quality management such as the study of the pollutants dispersion in urban environments, the validation of existing dispersion models and the estimation of EFs,

- Propane contained in LPG was used as a passive tracer. This choice resulted to be a good alternative to other common tracer substances. Propane offers several advantages to traditional tracers: It is easily available, low price, non-reactive, negligible global warming potential, and easy to detect with commercial on-line gas chromatographs.

- The HCMC tracer experiment is the longest tracer study developed until now at roadside level. This was possible thanks to the advantages offered by the passive tracer selected and to the use of the on-line gas chromatograph.

- The results of the tracer experiment were also used to study the dispersion of pollutants in the street canyon. Results shows that wind speed and wind direction play an important role in the pollutant dispersion. The highest tracer concentrations were observed at the leeward side of the street when wind is perpendicular to the street axis. This behavior is called street canyon effect and it has been observed in other street canyon studies. These results are important for the air quality assessment because they prove the relevance of these meteorological parameters on the air pollution.

- The main use of the tracer emission system is to estimate the EFs. An alternative technique was developed to estimate the EFs. The results of the tracer experiment were used to calculate the dispersion factors and afterwards these dispersion factors were used together with road traffic counts and the pollutant measurements to estimate the EFs. In the case of HCMC, it was necessary to assume that the finite emission line placed in one site of the street represents the emissions produced by traffic in the entire length and width of the street and therefore there was an error associated to this assumption. A Computational Fluids Dynamics model (CFD) was used to calculate this error and to correct the estimated EFs. Results showed that the

corrected EFs are within the same range of the EFs reported in other studies. The proposed methodology results in an interesting and promising alternative to estimate the EFs.

Finally, the proposed method to estimate the EFs was critically evaluated by means of the Computational Fluids Dynamics Model (CFD) MISKAM. The results of the tracer experiment were used to evaluate the performance of the CFD model. Different alternatives to improve the propose methodology were assessed with the model. The conclusions are:

- The CFD model MISKAM is able to reproduce quite well the average behavior of the tracer in the street canyon. The model reproduces the same average trends and levels of the observations in most of the cases. There is an outstanding performance when wind is perpendicular or oblique to the street axis but the model underestimates the concentrations when wind is parallel to the street axis. This underestimation is attributed to the size of the chosen simulation domain.

- The CFD model was used to critically evaluate the error produced by the source configuration used in HCMC. Results show that the position of the source produces larger errors than the length of the emission line. The simulations show that using the same emission line length used in HCMC but placed in the center of the street would produce even better results than a larger emission line but placed in one side of the street.

- The CFD model was also used to find the tracer emission source configuration that better approximates to the vehicle emissions and that may be used in future studies to calculate more accurately the EFs. Results showed that a 200 m line placed in the center of the street would represent very well the vehicle emissions.

- Finally, the proposed methodologies and especially the combination tracer experiment / CFD model shows to be a very promising tool for the air quality management.

In summary, the developed methodologies serve different purposes at the same time and their use can provide useful information for the urban air quality assessment. That results interesting from the financial point of view. Concentrations of pollutants are determined at roadside level, as well as their evolution in time, this information can be used for exposure studies. At the same time, EFs can be determined under real urban conditions. Results from the tracer study can be used to validate dispersion models which in turn can be used in the future to evaluate abatement strategies for such streets.

6.2. RECOMMENDATIONS FOR FUTURE RESEARCH WORKS

The experience gained during the development of this PhD thesis let to formulate some recommendations for future research works. Recommendations are:

- It is necessary to measure a larger number of pollutants. As it was stated in the second chapters of this thesis, hundreds of pollutants are released to the atmosphere but in developing countries only major air pollutants are monitored. In the case of HCMC we only measured VOCs from $C_2 - C_6$, in this city it is necessary to measure at least the VOCs from $C_6 - C_{10}$. These pollutants are important not only because some of them produce serious health effects but also because they play a key role in the atmospheric chemistry. It is also necessary to find the sources of these pollutants.

- The HCMC tracer experiment was carried out in an urban street canyon. It would be very interesting to develop the tracer experiment in an open urban street. These results could be used to estimate the EFs under many different urban conditions and also to validate existing models developed to simulate the dispersion in such environments.

- The results of the tracer experiment developed in HCMC were used to validate the CFD model MISKAM. It would be very useful to evaluate other CFD models or even the parametric models. It would be very useful also to develop an inter comparison of models. The existing database is available to anyone interested.

- During the estimation of the EFs it was found that the pollutants background concentrations had an effect on the accuracy of the calculations. Therefore it is recommended either to develop the measurements in a place with significant traffic emissions or either to measure the background.

- The results of the CFD simulations showed that a 200 m emission line placed in the center of the street would represent very well the emission produce by road traffic. It would be very interesting to experimentally test this tracer source configuration and to use the results to estimate the EFs in a city.

- The CFD model MISKAM was able to reproduce very well the average behavior of the tracer in the street canyon despite it doesn't consider thermodynamic processes like energy transformation at the surface of the road, neither walls, roofs of buildings, nor thermal dispersion, buoyancy or water balance. In the case of HCMC, the CFD model produced very good results without considering this parameter on the simulations. It will be necessary to develop similar studies in other cities of the world and to evaluate if it is really necessary to include this parameter on CFD models.

This thesis contributed to improve our understanding of the urban air pollution problems in developing countries. Despite the developed methodologies were initially conceived for cities of the developing world, they can be also used in developed countries. These methods may be an additional tool for the correct assessment of road traffic emissions in such countries. As it is known, many cities of developed countries also face air pollution problems and in many cases the road traffic emissions are still uncertain.

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2007 - Aujourd'hui	Professeur invité à l'**université de los Andes** à Bogota, Colombie. Chargé de cours en *Thermodynamique*. dans le "Departamento de Ingeniería Civil y Ambiental" .
2001 – 2007	Ingénieur et Chercheur à l'**université de los Andes** à Bogota, Colombie. Durant cette période j'ai effectué plusieurs études de qualité de l'air pour la ville de Bogota. J'ai également participé au développement d'un système d'aide à la décision en matière de gestion de la qualité de l'air de la ville de Bogota.
2003 – 2005	Professeur adjoint à l'**université de los Andes** à Bogota, Colombie. J'ai dispensé environ 500 h de cours à des publics très variés: étudiants en Bachelor et Master en Sciences et Ingénierie de l'Environnement.

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Belalcazar, Luis; Clappier, Alain; Blond, Nadege; Flassak, Thomas ; Eichorn, J; 2009. Impact of different source configurations on the estimation of vehicle emissions factors from tracer studies. Submitted to Atmospheric Environment.

Belalcazar, Luis; Ho, Minh-Dung; Fuhrer, Oliver; Clappier, Alain; 2008. VOCs and other pollutants associated to motorcycles in the atmosphere of Ho Chi Minh City, Vietnam. Submitted to Atmospheric Research.

Zarate, E; Belalcazar, L C; Clappier, A; Manzi, V; Hubert Van den Bergh; 2007. Air quality modeling over Bogota Colombia; combined techniques to estimate and evaluate emission inventories. Atmospheric Environment, Vol. 41, 6302-6318.

Manzi, Veronica; Belalcazar, Luis; Giraldo, Eugenio; Zarate, Erika; Clappier, Alain; 2002. In Spanish "Estimación de los factores de emisión de las fuentes móviles de la ciudad de Bogotá". Revista de Ingeniería Universidad de Los Andes., Vol. 18.

Hernandez, A; Belalcazar, Luis; Rodriguez Manuel; Giraldo, Eugenio; 2002. Retention of Granular Sludge at High Hydraulic Loading Rates in an Anaerobic Membrane Bioreactor. Water Science and Technology. Vol. 45(10), I 10, pp 169-174.

LANGUES

Espagnol: Langue maternelle

Anglais: Très bonne maîtrise de l'oral et de l'écrit (publications et conférences)

Français: Très bonne maîtrise de l'oral

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