

TRF-TRAFFIC EMISSIONS

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TRAFFIC EMISSIONS

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LISBON AIR QUALITY – EVALUATING TRAFFIC HOT-SPOTS

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ABSTRACT

This paper presents the results from air quality campaigns carried out in two Lisbon traffic hot-spots (*Entrecampos* roundabout and *Liberdade* avenue), aiming to define a more accurate and appropriate location for the referred traffic monitoring stations, which are intended to be representative of the near-kerbside environment.

The campaigns were performed in spring and summer of 2004 with an air quality mobile laboratory equipped with instruments for the continuous measurement of NO_x, CO and PM₁₀. The monitored concentrations were correlated with traffic counts and wind direction. The results indicated that the mean variation of particle and gaseous concentrations closely followed the traffic pattern. This relationship was not evident for wind direction, probably due to the multiplicity of road contributions. Comparing with the air quality legislation, the results show its fulfilment, except for PM₁₀, which often exceeded the limit-value.

In order to evaluate and simulate the flow and dispersion fields of air pollutants emitted by road traffic, a computational fluid dynamics (CFD) model was applied. The simulation of wind and PM₁₀ dispersion fields for a 31 hours period was performed on a calculation domain centred at “Liberdade” Avenue. This domain is characterised by the presence of a group of buildings, streets and trees that result on a high geometrical complexity. Air quality data from the monitoring stations were used for the validation of the modelling results. Finally, this work stresses the importance of accounting for additional PM₁₀ contributions, as the non-exhaust road traffic emissions and the urban background concentrations.

Key Words: road traffic emissions, air quality, urban area, measuring campaigns, CFD model

1. INTRODUCTION

Nowadays, the whole world largely depends on individual mobility, which has contributed to a general increase in comfort standards. However, the air pollution caused by vehicles exhaust emissions has shown negative environmental and health outcomes. The rapid demographic growth of the last decades has increased the concentration of human populations in cities. As a consequence, the total emissions from road traffic have risen significantly, assuming the main responsibility for the

disregard of air quality standards (EEA, 2001), namely in the Lisbon city (Ferreira *et al.*, 2000).

The implementation of the European Framework Directive 1996/96/EC, which defines the pollutants to be measured and the systems to be used for those measurements, resulted in the development of *daughter* directives (including Directives 1999/30/EC and 2000/69/EC) that set the maximum levels for specific pollutants, which were transposed to the Portuguese Law through the Decree-Law n° 111/2002, from the 16th of April.

During the last years, several directives were adopted and plans had to be established with the aim of controlling air quality. Also, there has been an improvement on fuels quality and treatment technologies, with visible results in the improvement of air quality. However, urban environments continue to congregate the inherent problems to the major concentration of people, inappropriate urban planning, growing of the private transportation – with implications in the environmental problems of the city.

An automatic network consisting of seven stations monitors the ambient air quality in Lisbon, providing hourly data on the main atmospheric pollutants. The stations are distributed throughout the city in order to assess the atmospheric pollution in representative areas. Two of them, located at “Liberdade” Avenue and “Entrecampos”, could be regarded as typical urban traffic stations. During the last years, and especially in “Liberdade”, the daily limit value for PM₁₀ was exceeded more than 35 times per year. In this sense, one of the aims of this study is to analyse the location of the urban air quality stations in “Liberdade” and “Entrecampos” through the collection of data on air pollutants concentrations, meteorology and traffic fluxes. On the other hand, the current paper is focused on a modelling approach, complementary to the field campaign, for assessing the PM₁₀ air quality levels in a domain centred at “Liberdade” Avenue for a 31 hours period.

During the last years, Computational Fluid Dynamics (CFD) models have proved to be a reliable tool for the simulation of wind and dispersion fields in complex geometries, which are typical in urban areas, assuming an important role on scientific research, policy support and regulatory purposes. The development of powerful numerical codes, supported by the constant increase on hardware performances, has largely contributed to the increase of modelling accuracy. The possibility of knowing the value of any variable related with the mean and turbulent flow or the associated dispersion of an air pollutant in any location of a given tridimensional (3D) study domain is, in fact, one of the main advantages of this technique. However, measurement and modelling are complementary methodologies, in particular due to the fact that a great effort on increasing the performance of models towards the simulation of PM₁₀ is still needed, namely through the development of accurate methodologies for accounting for the contribution of the non-exhaust road traffic emissions and the urban background concentrations.

The overall goal of this work is to contribute to a better understanding of the air quality problem in urban traffic hot-spots by performing measurements and modelling studies of atmospheric pollutants concentrations in Lisbon.

2. MEASUREMENT CAMPAIGNS

2.1 “Entrecampos”

The choice of the sampling points took into account the North and South extremities of “Entrecampos” looking forward to be the most representatives of population exposure to pollutants levels originating from the nearest traffic ways: the roundabout itself; “Forças Armadas” Avenue (West); “Estados Unidos da América” Avenue (East); “República” Avenue (North and South). The sampling point related to the measurements carried through 15 and 21st October 2004 is close to the Air Quality Station (AQS) of “Entrecampos”, continuously measuring the same pollutants of the mobile unit of air quality (SNIF air lab).

Figure 1 presents the diagrams with the location of the campaign sampling points.

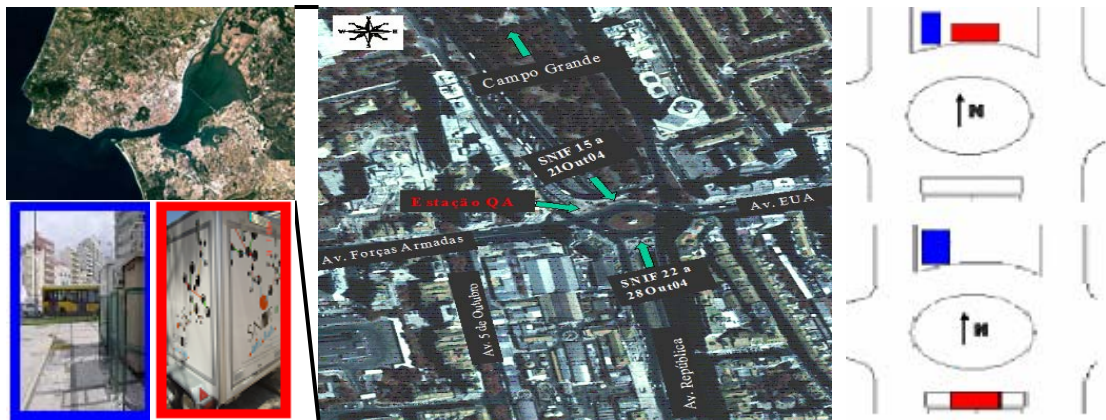


Figure 1 – SNIF air lab (red border); “Entrecampos” AQS (blue border); Lisbon (top and left) and “Entrecampos” (centre) orthophotomaps; SNIF location (red) and “Entrecampos” AQS (blue) in 15th to 21st October (top and right) and 22nd to 28th October (down and right).

All the NO₂ and CO results fulfil relevant norms, established in the National Decree-Law n°111/2002, 16 April. The mean values registered for PM₁₀, measured especially at the SNIF, exceeded the limit-value plus the margin of tolerance for the year 2004 (55 µg.m⁻³). The meteorological data (10 m high) were recorded at the Portuguese Meteorological Institute, located near the Lisbon airport, a few kilometres away from the measurement spot. Figure 2 presents the mean values obtained in the 2 weeks campaign (15-21 and 22-28, October).

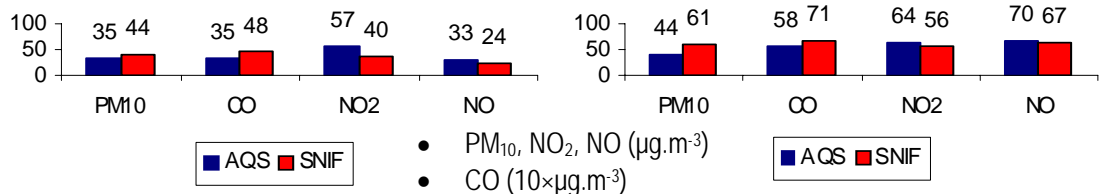


Figure 2 – Mean values of PM₁₀, NO₂, NO (µg/m³) and CO (10×µg/m³) obtained at the AQS and SNIF (left: October, 15 – 21; right: October, 22 - 28).

As shown, there was a consistent increase on the pollutants concentrations, at the AQS and SNIF, from the first to the second week. Concerning the meteorological parameters, there were no significant differences between both periods. The mean concentration values at the urban background stations (“Olivais” and “Loures”), increases comparing the first week to the second: 10-20% on PM₁₀, 34-104% on NO₂ and 7-82% on CO. There were also increases at AQS and SNIF: 26-39% to PM₁₀, 13-40% to NO₂, and 47-66% to CO. So, according to the meteorological data and from the results for the several pollutants measured at those stations, we can conclude that this phenomenon can be related with an increase of pollution, on a regional level, with consequent local effects.

During the campaign the five mentioned wind directions represent about 90% of the measured hours, while the three left wind directions (North, West and Northwest) had a rare occurrence. Consequently, the corresponding concentration values were unimportant and the respective averages subjected to enormous errors that could make difficult the analysis of the other results.

Figure 3 presents the hourly average results obtained at the AQS and SNIF; the percentage of occurrence of each wind direction and intensity (m.s⁻¹); and the traffic flux.

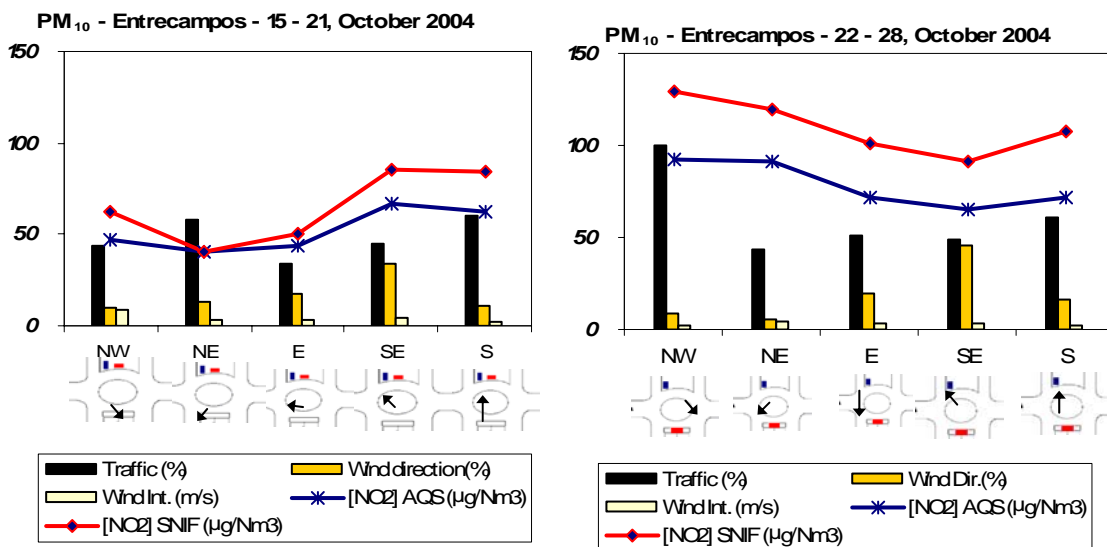


Figure 3 – Mean values of PM₁₀ obtained (Left: October, 15 – 21; Right: October, 22 – 28) at the AQS and SNIF, according to the wind direction; % of prevalence according to wind direction; mean relation of the traffic flow observed for each wind direction; and wind intensity (m.s⁻¹).

From the data collected some conclusions can be taken:

- As expected, the highest PM₁₀ concentrations were recorded at SNIF, when compared with the AQS. Similar values were uniquely observed when wind direction was from NE (1st week). From this direction, the air comes from the

North entry of the “Entrecampos”’s tunnel and from the woody area that extends from “Entrecampos” to “Campo Grande”;

- during the two weeks, the highest concentration levels correspond to wind directions favourable to draining of air masses in the traffic zone, concerning the sampling points: 1st week – SNIF registered higher values of Southeast wind; 2nd week – in a opposite point in the round, higher values of NW and NE wind were registered.

2.2. “Liberdade” Avenue

This campaign took place at the “Liberdade” Avenue, in 16th to 21st May 2004. The obtained results in SNIF and AQS (see figure 4) for NO₂ and CO fulfil the boundary-values stipulated in the national law n°111/2002. The mean values of PM₁₀ measured, especially at the SNIF, exceeded the limit-value plus the margin of tolerance for the year 2004 (55 µg.m⁻³).

Figure 4 presents the mean values obtained at the 2 sampling sites (16th to 19th and 19th to 21st May 2004), related to the hourly average values of each pollutant:

- NO and NO₂ were not measured in the first campaign period, due to problems with the analyser;
- the pollutants concentrations obtained during the first period at the SNIF were higher than those measured at the AQS. The SNIF was situated between the main traffic way of “Liberdade” Avenue and the AQS, registering a higher influence of traffic. These data showed that human exposure can be worse than expected;
- during the second measuring period, the opposite occurred: the values obtained at the AQS were always higher than SNIF’s. There was an almost null difference concerning CO. During the second measuring period the SNIF was more distant than the AQS from the main way of “Liberdade” Avenue, and so, under less traffic exposure and particle resuspension. These facts could influence positively the obtained results.

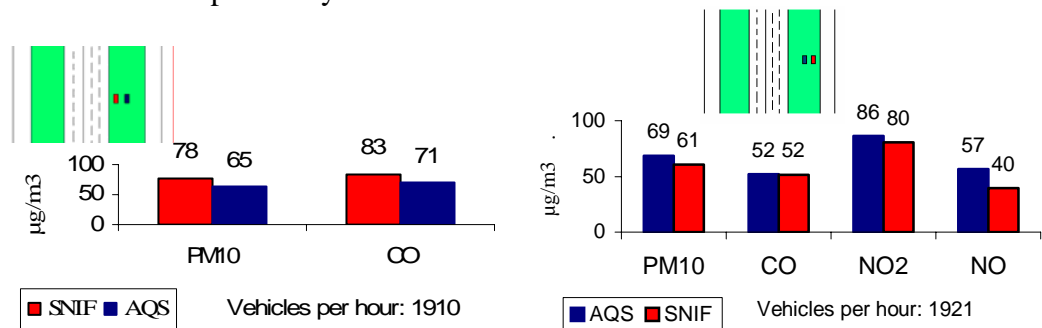


Figure 4 – Mean values of PM₁₀ (µg.m⁻³), CO (10×µg.m⁻³), NO₂ (µg.m⁻³) and NO (µg.m⁻³) obtained at the AQS (blue) and SNIF (red), between 16th to 19th May (left) 19th to 21st May (right); and hourly mean traffic flux.

3. MODELLING APPLICATION

The hourly simulations of PM₁₀ dispersion were performed for the period between 17h on the 19th of May and 24h on the 20th of May 2004. The methodology beyond the developed work was based on the application of the commercial CFD model FLUENT (Fluent Inc., 2003), version 6.1.18 for Unix platforms, previously applied by the authors (Amorim *et al.*, 2005) with successful results for the simulation of CO dispersion in a typical urban area. In the core of the solver, the model applies the Reynolds-averaged Navier-Stokes (RANS) equations for solving the mean flow, while all the scales of turbulence are modelled through the application of the $k-\varepsilon$ model. Both flow and dispersion fields were obtained applying an Eulerian approach. The selected domain (see Figure 5), comprising an area with 700 × 700 (L × W) m², was centred at “Liberdade” Avenue. In order to deal with the high geometrical complexity of buildings an unstructured mesh was created.

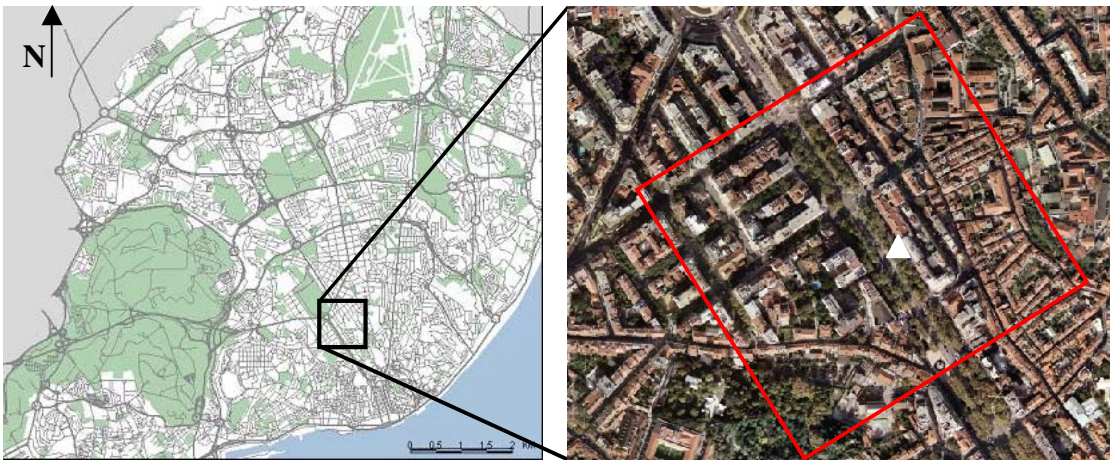


Figure 5 – Map with the location of the simulation domain of “Liberdade” Av. [adapted from <http://lisboainteractiva.cm-lisboa.pt/>]. The location of the AQS is represented by the white triangle.

The densely foliated tall trees that flank both sides of the Avenue along its entire length were digitally originated as 3D elements with a porosity defined according to a sink-term (S_i) added to the momentum transport equation. This term represents a simple parameterisation of the loss of wind speed due to pressure and viscous drag forces exerted by leaves and branches, and is expressed in Einstein notation as follows:

$$S_i = -C_0 |v|^{C_1} = -C_0 |v|^{(C_1-1)} v_i \quad (1)$$

where v is the wind velocity at that specific computational cell and C_0 and C_1 are empirical coefficients tuned to 10 and 1, respectively. Although empirical, this approach allows to obtain accurate results when compared to a more realistic vegetative canopy model already tested (Amorim *et al.*, 2005).

The hourly mean values of road traffic emissions of PM₁₀ were estimated through the application of the Transport Emission Model for Line Sources (TREM) (Borrego *et*

al., 2003 and 2004), using as main input the vehicles counting data with distribution by classes.

In figure 6, examples of PM₁₀ concentration fields for four different wind directions (in meteorological coordinates) are shown: 350° (18h on the 19th), 24° (1h on the 20th), 159° (14h on the 20th) and 216° (16h on the 20th). As can be seen, there is a clear accumulation of PM₁₀ along the Avenue, with the formation of some hot-spots. The location of those higher concentrations shows that the dispersion pattern is highly influenced by the wind direction.

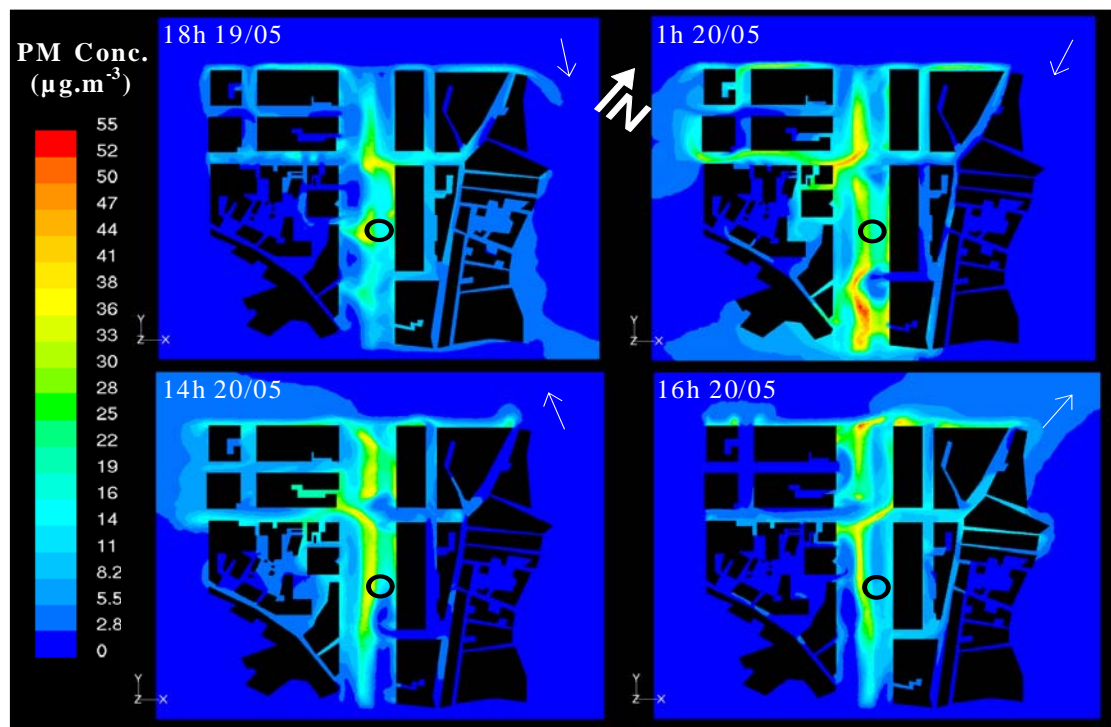


Figure 6 – 3 m high horizontal PM₁₀ concentration fields corresponding to four distinct wind directions. The location of the AQS is represented by the black circle. Vectors indicate wind direction angle.

In figure 7, the temporal variation of the hourly mean values of wind velocity, PM₁₀ emissions and PM₁₀ concentrations in ambient air are presented.

One of the conclusions that can be taken from the analysis of figure 7 is that PM₁₀ concentrations simulated by FLUENT (black solid line), with the consideration of the direct exhaust pipe emissions only, are significantly inferior to the measured ones (grey solid line). Therefore, a methodology was applied in order to consider the influence of the non-exhaust traffic related PM₁₀ emissions, which include brake-wear, tire wear and resuspension of loose material on the road surface.

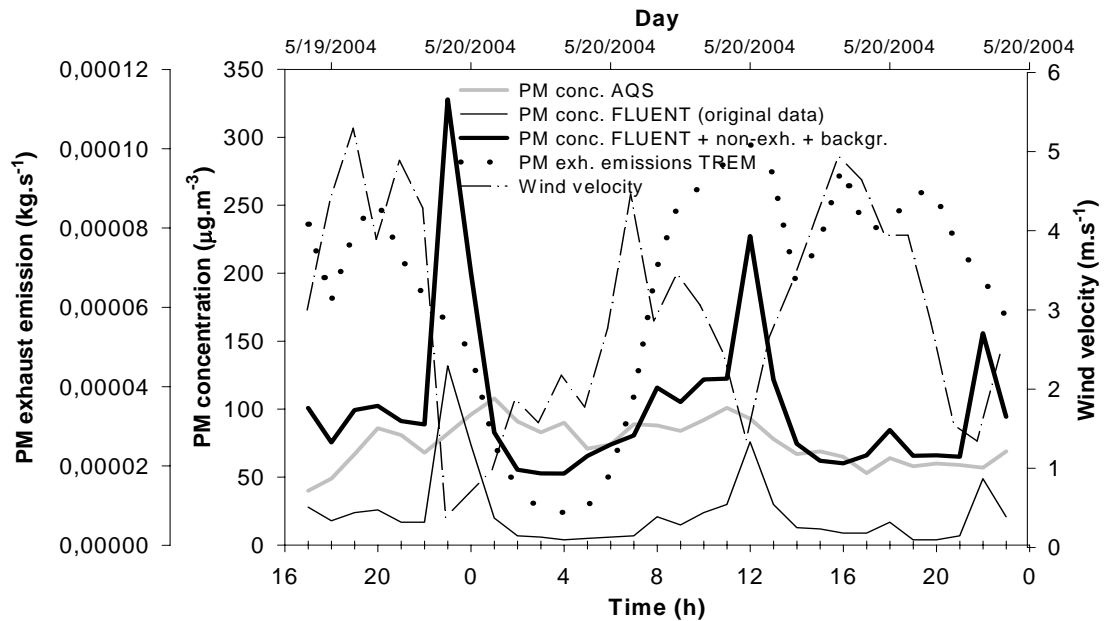


Figure 7 - Evolution with time of the mean hourly values of wind velocity ($\text{m}\cdot\text{s}^{-1}$) at the inlets boundaries, PM_{10} road traffic emissions ($\text{kg}\cdot\text{s}^{-1}$) estimated by TREM for the “Liberdade” Av., PM_{10} concentration ($\mu\text{g}\cdot\text{m}^{-3}$) measured in the AQS and simulated by FLUENT (original data and with the consideration of non-exhaust emissions and background concentrations) for the same location.

The PM_{10} emission factor of this non-exhaust contribution ($EF_{\text{non-exhaust PM}_{10}}$, in $\text{g}\cdot\text{km}^{-1}$) was calculated through the following equation:

$$EF_{\text{non-exhaust PM}_{10}} = 55.92 \cdot v^{-0.82} \cdot (sL/2)^{0.65} \cdot (W/3)^{1.5} \quad (2)$$

where, sL is the road surface silt loading, defined as $0.2 \text{ g}\cdot\text{m}^{-2}$ (value proposed by Düring *et al.* (2002) for a “good” surface urban street), v is the average vehicle speed, equal to $50 \text{ km}\cdot\text{h}^{-1}$, and W is the average vehicle weight of the fleet mix (1.1 t for cars, 1.9 t for light duty vehicles and 9 t for trucks, according to Düring *et al.* (2002)). This equation is based on the EPA (2003) model for the estimation of the dust emissions from dry paved roads, to which a speed component was added according to the methodology developed by Tchepel (2003).

The minimum and maximum quotient between total emission and exhaust pipe emission were, respectively, 1.9 at 1h and 4.4 at 6h, which are in agreement with the values found by Düring *et al.* (2004) for some motorways and major arterial roads in Germany, indicating that the non-exhaust contribution can be even greater than the direct exhaust one.

The PM_{10} background concentration was obtained as a mean value for two urban background air quality stations, “Loures” and “Olivais”, which are located at, respectively, N and NE of the domain. For this reason, this methodology is expected to give more accurate results for wind directions in the quadrant N/NE. Minimum

and maximum values of this background concentration are, respectively, $27.5 \mu\text{g}\cdot\text{m}^{-3}$ at 17h on the first day and $62.5 \mu\text{g}\cdot\text{m}^{-3}$ at 9h of the second one.

With the inclusion of these additional terms a much better agreement is found between simulated and measured values (solid bold line versus solid grey line, respectively). As can be seen, three distinct peaks are found which largely surpass the measured concentrations. These discrepancies correspond to the hourly simulations of 23h on the 19th and 0h, 12h and 22h on the 20th of May. These four hourly periods for which the difference between measured and simulated values is greater are, in fact, a result of the extremely low wind velocities (less than $1 \text{ m}\cdot\text{s}^{-1}$ at 10 m in height) registered. Moreover, the vegetative canopy contributes to an even higher reduction of the wind speed at the location of the AQS. The sensitive analysis applied to the 23h on the 19th showed that increasing the wind speed from the original 0.4 to $2 \text{ m}\cdot\text{s}^{-1}$ and $5 \text{ m}\cdot\text{s}^{-1}$ a decrease of, respectively, 78 and 91 % in the PM simulated value is obtained.

The Normalized Mean Squared Error (NMSE) for the air quality values simulated by FLUENT with the consideration of the non-exhaust and the background contributions is 0.487. However, neglecting the values corresponding to the observed peaks, this statistical parameter decreases to 0.086, indicating a very good modelling performance (significantly better than 2.061, for no non-exhaust and background additional inputs). According to the Directive 1999/30/EC there are not yet data-quality objectives for assessing the uncertainty associated to PM_{10} modelling in terms of hourly or daily averages. New legislation is therefore necessary to fulfil this gap.

4. CONCLUSIONS

The air quality characterization in “Entrecampos” and “Liberdade” Avenue, that involved the results of the mobile laboratory (SNIF) and the AQS, allowed to conclude that:

- NO_2 and CO results fulfil the respective air quality norms, stipulated in the Decree-Law n. ° 111/2002, of 16th of April. Some of the PM_{10} daily average values, specially in the SNIF, exceeded the respective limit value plus the margin of tolerance;
- it is still possible to verify that the SNIF had normally higher values than the AQS, probably because of the greater proximity to the road;
- it is clear the influence of wind direction in the pollutants levels, namely for PM_{10} , and specially in “Entrecampos”;
- the proximity to the traffic way of the “Liberdade” Avenue, generally represents greater concentration values, to the exception of the CO, whose trend is not so evident;
- the results indicated that the mean variation of particle and gaseous concentrations followed the traffic flow rate, but not so evidently in relation to wind direction, probably due to the multiplicity of road contributions.

The performed simulations showed that current methodologies for PM_{10} dispersion modelling allow to obtain acceptable results on air quality assessment within

complex urban areas. Nevertheless, the importance of the contributions of non-exhaust road traffic emissions and urban background concentrations on total PM₁₀ concentrations fully justifies the need of improving the accuracy in the quantification of these additional sources of particulates.

Further developments on this specific area of research should allow to contribute to the establishment of adequate data-quality objectives for assessing the uncertainty associated to PM₁₀ modelling established by the European Law.

The campaigns results and the performed simulations allow us to obtain important information for the development of adequate methodologies towards air quality evaluation. It may also be used to promote the necessary knowledge of the potential risk for human health in traffic hot-spots areas and to guarantee the existing fulfilment of the norms and legislative aspects.

5. ACKNOWLEDGMENTS

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BENZENE IN THE AIR OF ZAGREB, CROATIA – FIRST STUDY

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ABSTRACT

This paper presents the results of the first measurements of benzene concentrations in the air of Zagreb, Croatia. The samples were taken at two sampling sites and analysed every 15 minutes using automatic gas chromatographs. One sampling site was the national monitoring station located in a densely populated city centre with high dense traffic. The other sampling site was a regional monitoring station in the northern part of the city with small population density and low traffic. The measurements were carried out simultaneously at both sampling sites from May 19 to August 16, 2004. The results showed diurnal variations characteristic of urban areas and the influence of traffic, with higher values measured in the morning hours. Both sampling sites showed differences in benzene concentrations between working days and weekends. When comparing benzene levels between the two sampling sites it was found that benzene concentrations were lower in the northern part of the city throughout the sampling period.

Key Words: air pollution, sampling site, traffic.

1. INTRODUCTION

Benzene is a ubiquitous air pollutant, and the general population is permanently exposed to it. The International Agency for Research on Cancer (IARC) has classified benzene as a carcinogen in humans and there is no identifiable threshold below which there is no risk to human health (IARC, 1982; Benzene, WHO, 1993). According to the Directive 2000/69/EC of the European Parliament, the limit value for benzene concentration in ambient air is $5 \mu\text{g m}^{-3}$ as an annual average (Directive, 2000). In most European countries there is a network of stations for measuring ambient air pollution, including benzene concentrations in the air. There are also some studies (for example MACBETH programme) that were designed to quantify population exposure to ambient atmospheric benzene (Gonzalez-Flesca et al., 2000). Measurements of benzene concentrations in the air in Croatia have been started only recently. Current measurements are continuously conducted at two locations in Zagreb: at the national and regional monitoring station.

The aim of this paper is to present data of the first measurements of benzene concentrations in the city of Zagreb, Croatia and to evaluate if there is significant

difference between benzene concentrations measured at the two sampling sites with different traffic density.

2. MATERIALS AND METHODS

Measurements of benzene concentrations were carried out simultaneously at two sampling sites in Zagreb, Croatia from May 19 to August 16, 2004. One sampling site was the national monitoring station located in a densely populated city centre at the distance of approximately 20 m from two roads (crossroad) with high traffic density. The other sampling site was a regional monitoring station in the northern, residential part of the city with small population density and low traffic. That station was located at a distance of more than 200 m from the nearest road or parking lot. Elevation of the sample inlet was 2.5 m from the ground level at both sampling sites. The fifteen-minute samples were collected and analysed using automatic gas chromatographs (VOC71M, Environnement s.a., France; AirmoBTX 1000, Airmotec, Germany) with PID and FID detectors.

3. RESULTS AND DISCUSSION

The results of benzene concentrations are shown in Figure 1. The average mass concentrations and selected statistical parameters are shown in Table 1. Daily variations of benzene mass concentrations, measured over 90 consecutive days show corresponding variations at both sampling sites. Those variations mainly depend on weather conditions and they were identical at city centre and at the northern part of the town. One-way analysis of variance (ANOVA) shows significant difference ($P < 0.01$) in mass concentrations of benzene between the two sampling sites. Over the whole measuring period, higher values were measured at the city-centre sampling location, with the maximum daily average of $5.8 \mu\text{g m}^{-3}$. Both sampling sites were selected in order to avoid any effect of stationary industrial sources. Thus, the higher benzene concentrations at the city-centre sampling site are as expected, considering high traffic density of the area.

The Ordinance on Recommended and Limit Air Quality Values base on the Croatian Law on Air Quality Protection defines the following values for benzene:

- Recommended value = $2 \mu\text{g m}^{-3}$,
- Recommended value of 98th percentile = $5 \mu\text{g m}^{-3}$,
- Limit value = $5 \mu\text{g m}^{-3}$,
- Limit value of 98th percentile = $10 \mu\text{g m}^{-3}$.

All values are defined for 8-hour running averages (Law, 1995; Ordinance, 1996). Although the usual averaging period for categorizing the air quality of the area with respect to benzene involves one calendar year, our results show that the limit value of $5 \mu\text{g m}^{-3}$ (also given by the Directive 2000/69/EC of the European Parliament) was exceeded in the city centre on three occasions during the sampling period.

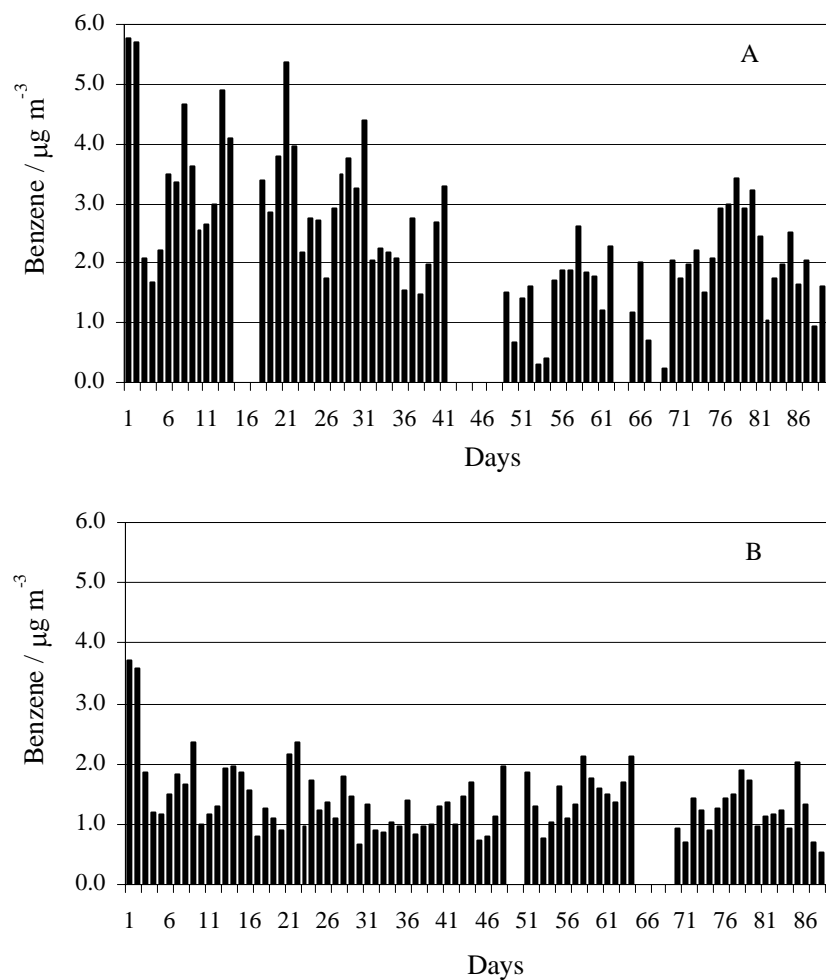


Figure 1. Daily variations of benzene concentrations measured in A) city centre and B) northern part of the town, in the period May 19 to August 16, 2004.

Table 1. Mass concentrations of benzene ($\mu\text{g m}^{-3}$)

Sampling site	N	C	SD	C_{\min}	C_{25}	C_{50}	C_{75}	C_{98}	C_{\max}
Center	77	2.5	1.2	0.2	1.8	2.2	3.0	5.5	5.8
North	82	1.4	0.6	0.5	1.0	1.3	1.7	2.8	3.7

N - number of samples

C_{\min} - minimum values

C_{75} - 75 percentile

C - average values

C_{25} - 25 percentile

C_{98} - 98 percentile

SD - standard deviation

C_{50} - median

C_{\max} - maximum values

Diurnal variations of benzene concentrations measured at the sampling sites located in city centre and in the northern part of the town are shown in Figure 2.

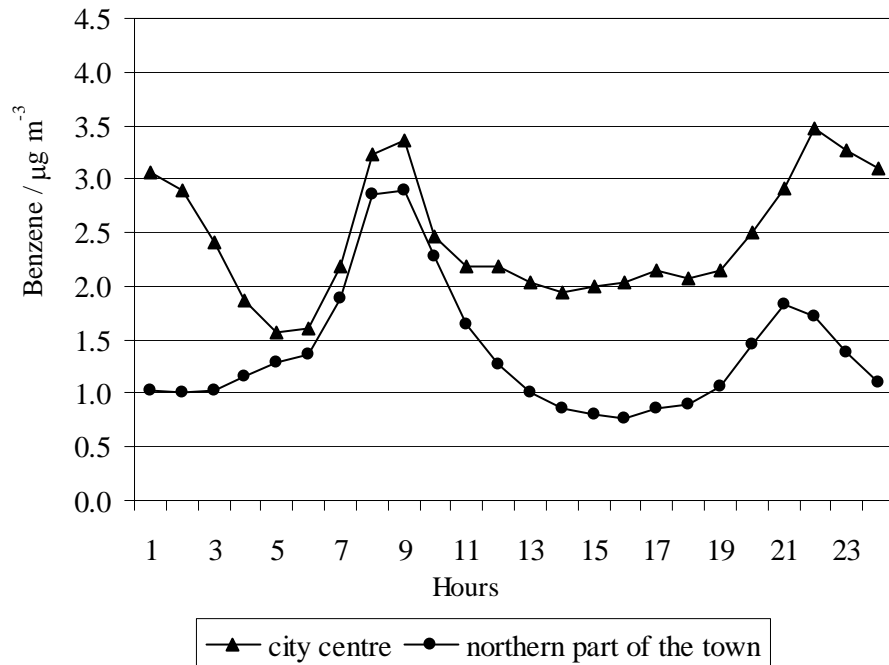


Figure 2. Diurnal variations of benzene concentrations measured at two sampling sites.

Trends are very similar at both sampling sites. Mass concentrations of benzene start to increase at 6 a.m. and they reach the daily maximum between 8 and 9 o'clock in the morning when most people start to work. Personal cars are still preferable instead of public transport, so morning traffic jams are very often. After 9 a.m. the benzene levels start to decrease. Another maximum, observed in the evening hours, can be explained in Figures 3 and Figures 4 with separate values for weekends (Saturday, Sunday) and working days (Monday to Friday). Differences between benzene concentrations measured on weekends and on working days in city centre are shown in Figure 3. Figure 4 presents diurnal variations on working days and on weekends measured in the northern part of the town.

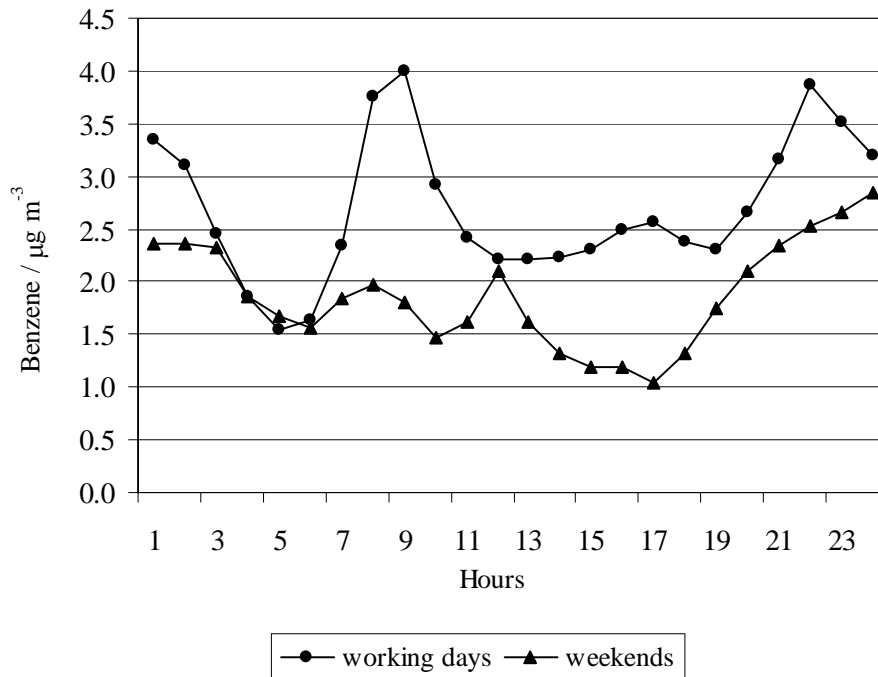


Figure 3. Average hourly values of benzene concentrations measured in the city centre.

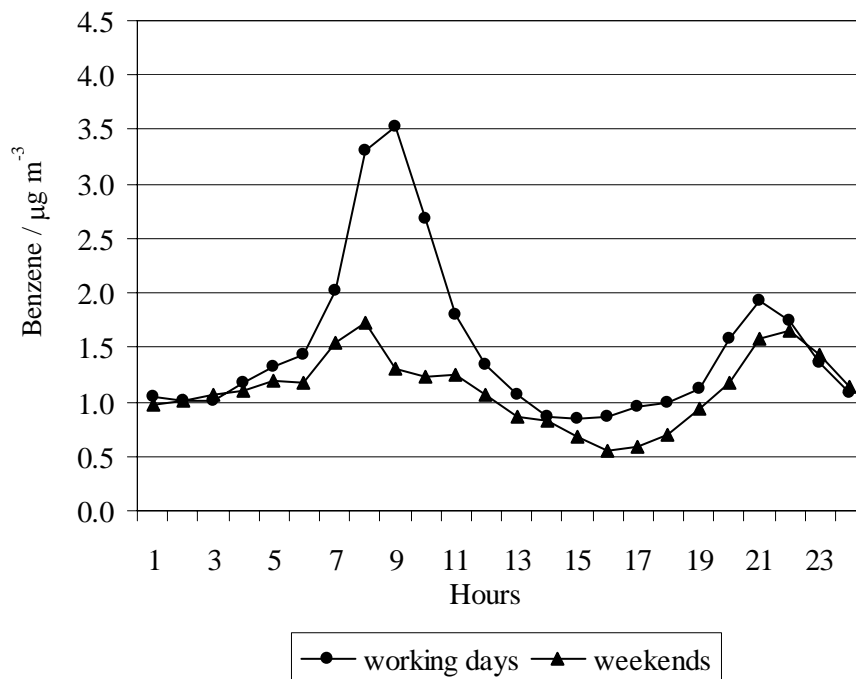


Figure 4. Average hourly values of benzene concentrations measured in the northern part of the town.

Diurnal variations of benzene concentrations between working days and weekends differ on both sampling sites. Different trends between sampling sites was also found. Elevated values were measured on working days, and also at the sampling site in city centre. The maximum daily values from 8 to 9 a.m. during the working days were not observed on weekends. The increase in benzene levels was observed in city centre in the evening hours, especially on weekends. All results indicate strong influence of traffic as a major source of benzene concentrations in the air of Zagreb, Croatia.

4. CONCLUSION

First measurements of benzene concentrations in the air of Zagreb, Croatia, are presented. Concentration levels of benzene showed significant difference between the two sampling sites with higher traffic-related values at sampling site in city centre. Limit value of $5 \mu\text{g m}^{-3}$ was exceeded on three occasions in city centre. Diurnal variations showed maximum values between 8 and 9 a.m. at both sampling locations. Benzene concentrations measured on working days and on weekends confirmed the influence of traffic.

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PREDICTING SHORT TERM VEHICULAR RELATED AIR POLLUTION FOR CORRIDORS: A CASE STUDY IN ISTANBUL, TURKEY

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ABSTRACT

This paper suggests a method by which a governmental agency can use short term traffic forecasting and air pollution modeling techniques to predict air pollution (i.e., CO, NO_x, particulate matter) for major corridors for one or two days in advance to provide a simple advisory system for decision-makers and the public. The authors propose the use of a Geographic Information System (GIS) integrated with a travel demand and an air pollution dispersal model. This system will first be tested on a corridor in Istanbul, Turkey. After the model has been tested for this corridor, the authors would discern how it could apply it for the major corridors in Istanbul. It is thought that this system will be a very useful device for decision-makers to warn the public about possible health hazards, but also to view alternatives to mitigate the effect of traffic pollution. The public, in turn, could lessen the situation by choosing alternative routes in predicted areas of high pollution or using public transportation.

Key Words: GIS, travel demand modeling, CO, NO_x, Air Pollution Dispersal Modeling.

1. INTRODUCTION

Air pollution related to vehicular traffic in corridors has been proven to have serious short and long term health consequences to the residents living near them (Krzyzanowski, 2005; Minnesota Center for Environmental Advocacy, 2003). Young children and the elderly are particularly vulnerable to this type of pollution. Educational institutions, social service agencies, and governmental buildings in densely urbanized areas are often placed immediately adjacent to congested arterials. The primary concern of governmental agencies has been the overall pollution of an urbanized area. For the most part, the consequences of traffic related air pollution in small areas where people may be at the greatest risk are ignored. It is estimated that more people die of traffic related air pollution than traffic accidents.

The key to being able to mitigate the effects of traffic related air pollution is adequate modeling tied to decision-making. If public officials can alert the public about severe traffic related pollution along certain corridors, then segments of the population such as the young or the elderly can stay indoors or refrain from serious physical exercise. In the long term, being able to model traffic related pollution would enable local

decision-makers to evaluate the effectiveness of mitigation strategies and better plan new urban development.

There is well developed body of research in air dispersal models related to vehicular traffic (Angelino et al., 1999; Dommen et al., 2003; Gulliver and Briggs, 2005; Jensen et al., 2001; Lin, and Lin, 2002; Masud, 1999; Maurizio et al. 1998; Reynolds and Broderick, 2000; Xia and Shao, 2005). This topic is complex because of the numerous variables such as the types of pollutants (i.e., CO, NO_x, particulate matter); the time of day; photo-chemical reactions; meteorological conditions; amount and composition of traffic flow; topography and heights of buildings and concentration (i.e. “the urban canyon effect”); and scale/resolution. Because of these numerous factors, the models developed for traffic related air pollution are data intensive and necessitate complex formulae. Nevertheless, the application of such models is limited in application. In addition, the effects of traffic related air pollution in urbanized areas in developing countries with high density, limited regulations and strained transportation infrastructure is very sparse.

The use of Geographic Information Systems (GIS) has proven to be effective in modeling and the monitoring of traffic related air pollution. GIS modeling is directly related to the linear aspects of traffic related air pollution. The use of raster models have also been use to model traffic related air pollution. The use of a GIS linked to the Internet to provide real time information about air pollution in several cities. The linking modeling and real time information for air pollution related to traffic is not an unrealistic extrapolation of the use of GIS and the Internet.

2. CASE STUDY AREA

The area selected for the testing the model is Istanbul, Turkey (see figure 1.) Istanbul has developed extremely rapidly over the last twenty years to become one of the largest cities in Europe (Ocakci, 2000). The population is estimated to be approximately fifteen million. Istanbul, like other high developing countries is the “primate city” for Turkey and has received the majority of growth. The growth has been haphazard with little or no planning. In the suburban areas, high rise developments are common contrasting with the relatively low rise development in the older center. With the rapid development, urban infrastructure such as roads and public transport systems are lagging behind the demand. Congestion is often severe along the major corridors

Istanbul growth has been fueled by the development of highways running east to west.

The linking of the European and the Asian side by two bridges has also been a major component in the growth of Istanbul. The growth has been high density along these corridors. There has few major arterials traversing Istanbul from north to south.

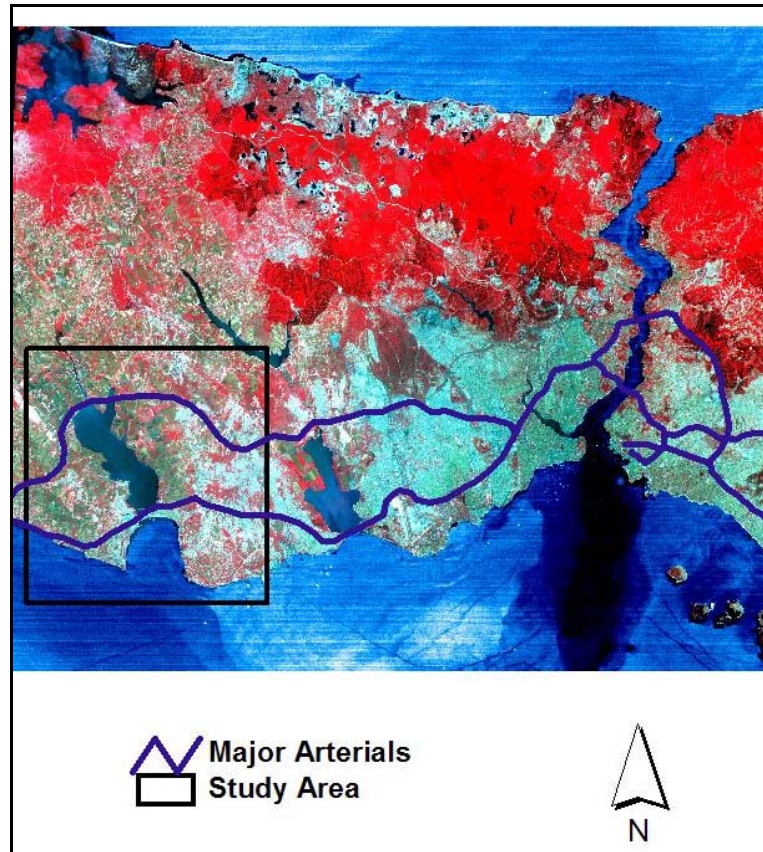


Figure 1. Istanbul urbanized area with Study Area

Istanbul has a different vehicle composition in the traffic flow than is found in developed countries such as Germany, the UK or the United States. There are a great many older vehicles that pollute more. Due to the lower automobile ownership, there is a large amount of buses (Ocakci, 2000). In addition, there is a large amount of trucks.

Because of the unbridled development, one often finds intense development directly adjacent to major corridors. This includes residential, commercial, and institutional land uses (i.e., schools, hospitals, governmental buildings etc.). It is obvious that residents and workers in these highly congested corridors are being exposed to significant traffic related air pollution. However, to the authors knowledge, local government is unaware of this problem and therefore do not have the necessary information to develop appropriate mitigation measures.

A corridor in Istanbul was selected to develop a short-term advisory system related to the levels of traffic related air pollution, specifically CO, NO_x and particulate matter (See Figure 2).

The corridor is located within the Büyükçekmece Municipality of the Greater Metropolitan Istanbul Area (Istanbul Büyükşehir Belediyesi.) It is a moderately

dense area and has a variety of land uses including institutional, commercial and residential.

At a later time, specific information such as land use using high resolution remote sensing images will be included in the GIS database. This could be used to identify which particular building would be affected by the air pollution

3. DESCRIPTION OF MODEL

The proposed pollution dispersion model (see figure 2) would integrate travel demand and meteorological models within a GIS to predict the effect of pollutants for a short time period—one or two days in advance. The travel demand model would be a key element in this system. It would include existing traffic counts, capacities, composition of the traffic (i.e., automobiles, trucks, buses etc.), and key traffic generators along the corridor.

Using the predicted volumes and vehicle composition, traffic related air pollutants would be estimated for the corridor using pollution factors suitable for Istanbul. It is estimated that the pollution would follow a Gaussian distribution along the corridor.

Concentrations would be estimated for a buffer area in the corridor for about two hundred (200) meters. The intervals would be spaced at twenty (20) meters. It would be estimated that the amounts of the pollution would be most concentrated along the immediate corridor with variations based on the predicted weather conditions. These estimates would be put in categories according to standards.

These levels would be displayed in a GIS according to their severity. This GIS display could be integrated with the Internet for informing decision-makers and the public about the risk of pollution for their particular area.

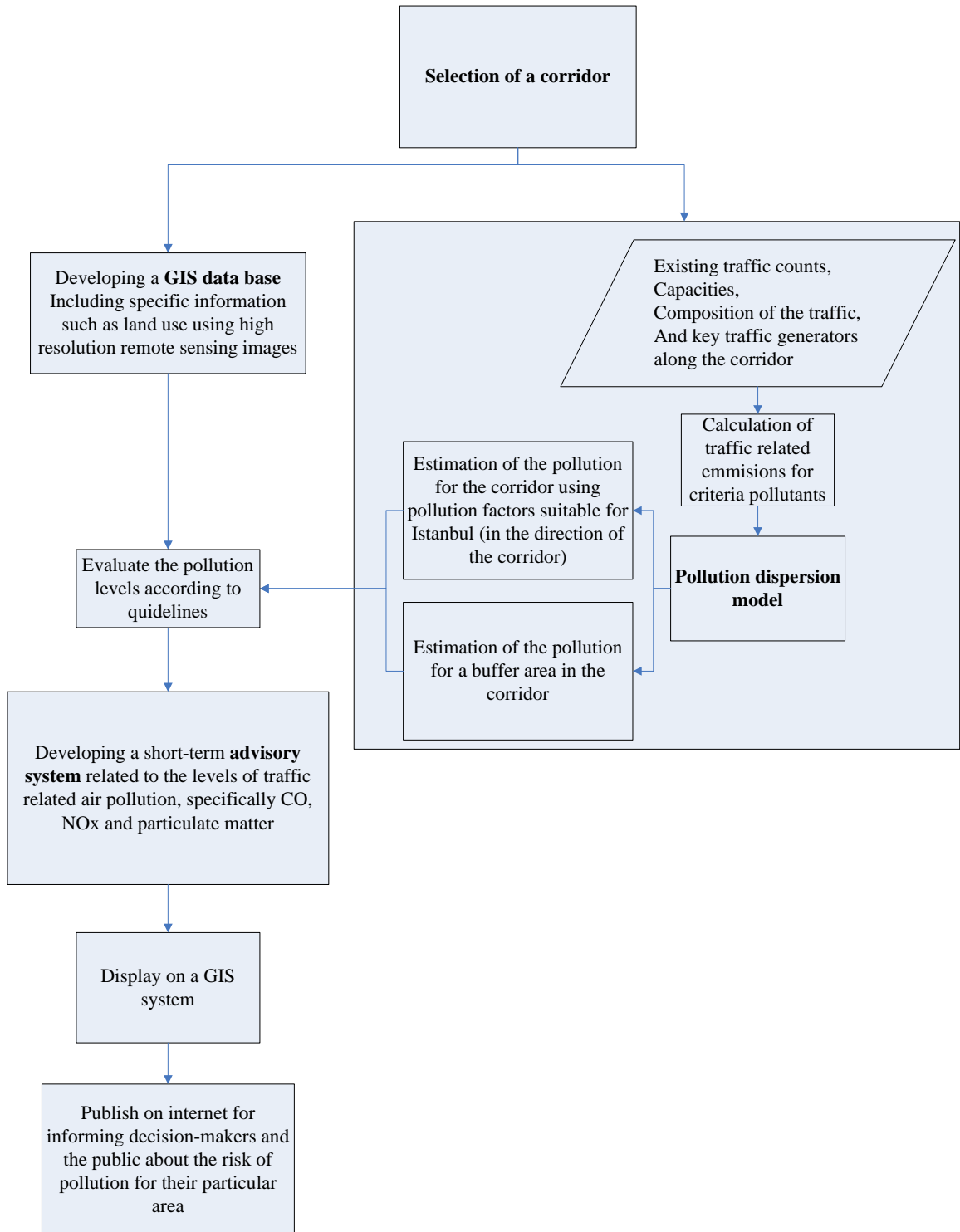


Figure 2. Description of proposed GIS based pollution dispersion model for corridors

4. RESULTS AND DISCUSSIONS

One of the most important tasks of these kinds of studies is the calculation of traffic related emission ratios and distributions of pollutants. As an application of this method, CO₂ was selected as parameter for this case study. CO₂ was selected for the test pollutant because it can be directly related to vehicle emissions. For evaluation of CO₂ over the selected corridor, a traditional Gaussian distribution model was used.

To simplify the results, it was assumed that meteorological conditions are stagnant (no wind direction and no air turbulence). The results are displayed by using a GIS system in the figures 3 and 4.

The hourly traffic data of selected area was estimated using existing traffic count data and visual observations at various places along the corridor. The volume of the traffic on the corridor changes from 50 car equivalents per/min to 300 car equivalents cars per/min. For the transformation of vehicles to car equivalents, we used the method suggested by Xia and Shao (2005).

The results of the model could be viewed in two different formats according to purpose: One format would be the specific aspects of the pollution distribution at high resolution, as seen in Figure 3. The other is the advisory system as illustrated in Figure 4. Figure 3 is the close up of the pollution buffers of the corridor, controlling for meteorological conditions.

The heaviest concentration would be in the buffers immediately adjacent to the arterial with lesser amounts in the outlying buffers. This concentration distribution would change depending on the particular day of the year. The resolution of such a buffer is important to determine particular land uses (such as schools, hospitals etc.) that may be adversely affected by the pollution.

The advisory map is a summary for decision-makers and the public (see Figure 4). There are some locations because of either steep slope, buildings or congestion caused by entering vehicles, which would be areas of high pollution. For example, Sector 5 is an area where are many buses stopping and causing congestion.

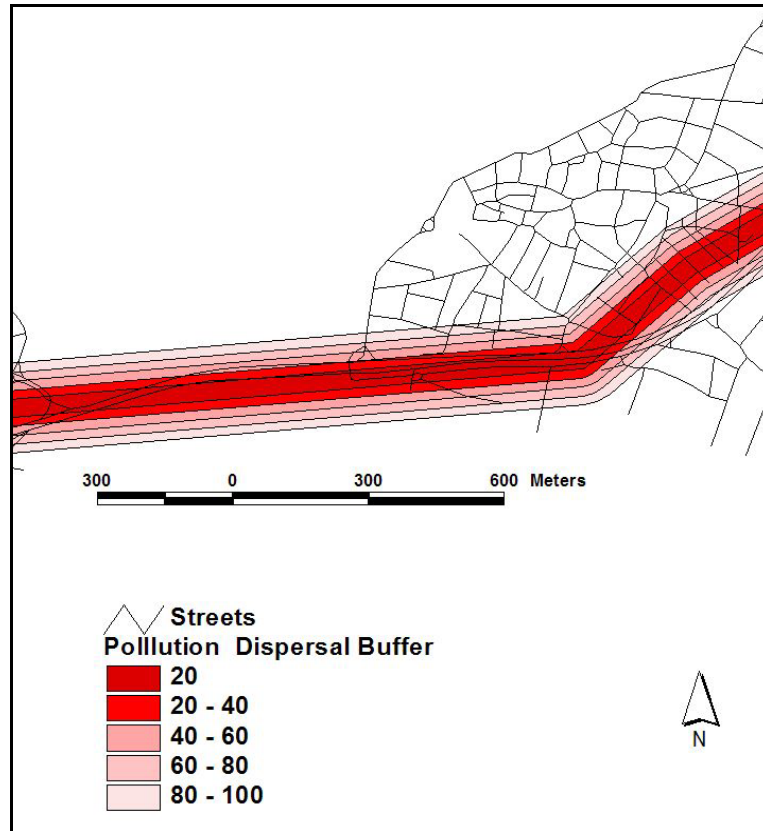


Figure 3. Close-up of CO₂ pollution dispersal buffer for the corridor under stagnant meteorological conditions (no wind direction and wind speed)

5. CONCLUSIONS

The paper suggests a method by which a governmental agency can use short term traffic forecasting and simplified air dispersal modeling techniques with weather forecasting for particular micro-climates for predicting air pollution measurement for a particular corridor for one or two days in advance.

The model is fairly complex due to the aspects of modeling correctly the amount of pollutants and the dispersal based on micro-climatic conditions. The authors will be developing the actual model so that the usefulness of such a system can be discerned.

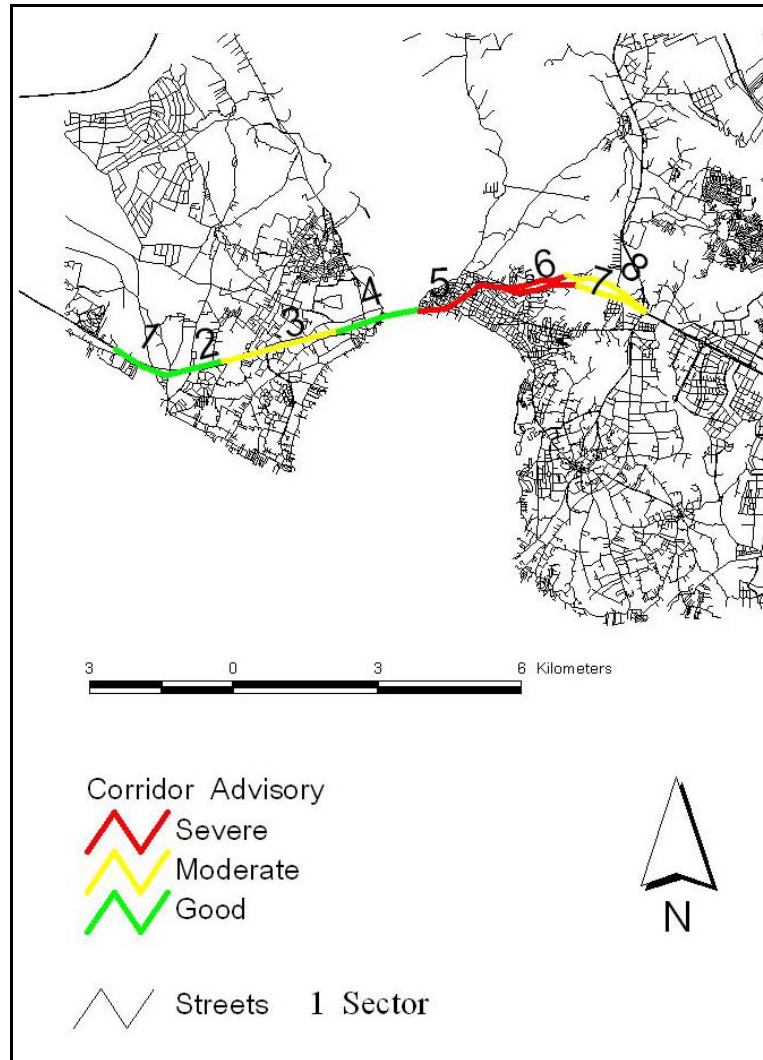


Figure 4. Sample Corridor Advisory System

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IMPROVED METHODOLOGY FOR ROAD EMISSIONS CALCULATION: THE CASE OF PORTUGAL

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ABSTRACT

A new national methodology to estimate road transportation emissions was developed with the main objective of improving emission estimates for those pollutants that are most dependent on the vehicle class and abatement technology. Emissions were estimated with a detailed level of disaggregation allowing the evaluation of measures aiming key source groups rather than road traffic in general. Those measures include reduction of speed limits, changes in fleet age or engine capacity, introduction of new technologies, among others. The methodology is coherent with the National Emission Inventory in terms of assumptions, activity data and emission factors.

Key Words: Road Transport, Emissions, Climate Change, Air Quality

1. INTRODUCTION

Emission estimates are required for various objectives. In first place, at national level, each country is required to submit emission estimates under the Convention on Long Range Transboundary Air Pollution (CLRTAP) and under the Framework convention on Climate Change (UNFCCC). The rapid increase in road traffic emissions in the last decade makes this sector one of the most important topics. In the other hand, the accomplishment of air quality at regional and city level involves the use of diagnosis and predictive models which require the elaboration of detailed emission inventories. Therefore, the emission inventories must be complete, reliable, and accurate.

Preferably the emission inventory for road sources should be based in knowledge of traffic flow and travelling conditions. However this information is usually limited to major roads. That is the case in Portugal where traffic monitoring is restricted to toll highways (about 2000 km in 2003), and some point survey in major non-toll highways and national roads. Given the incompleteness of these data, total emissions must be estimated from fuel consumption, i.e. coordinating a bottom-up approach together with a top-down approach. The proposed methodology to estimate

emissions from road transportation uses a combination of country-specific and international methodologies and emission factors.

Transportation emissions from the analysed countries are responsible for similar shares from overall emissions, with the sole exception of Norway. Moreover, the share of road transportation within transport emissions is very high (most values greater than 90%).

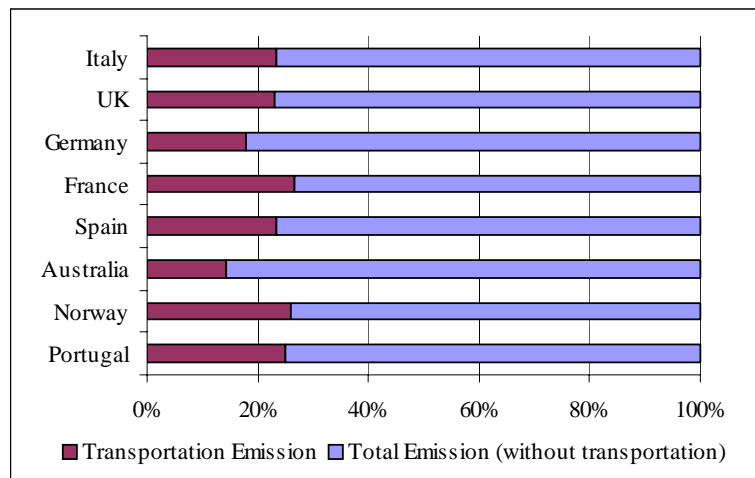


Figure 1. Share of transportation emissions (CO₂ equivalent) from the overall emissions (Source: UNFCCC, 2002).

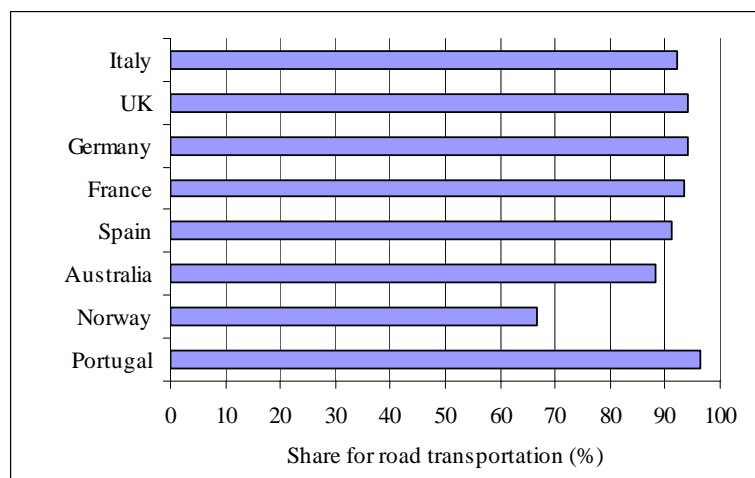


Figure 2. Share of road transportation emissions (CO₂ equivalent) from overall transport emissions (Source: UNFCCC, 2002).

2. METHODOLOGY

The model for exhaust emission estimate follows a country specific integrated methodology translated into a Visual Basic for Applications[®] (VBA) software tool linked with Microsoft Excel[®] spreadsheets. This model is compatible and based

extensively in EMEP/CORINAIR Emission Inventory handbook (EEA,2002) and reproduces emission factors that were derived from the COPERT III Computer programme to calculate emissions from road transport (Ntziachristos and Samaras, 2000). Most of the countries under the UNFCCC which submitted annual inventories of greenhouse gas emissions by sources and removals by sinks used a combination of country-specific and IPCC methodologies and emissions factors. National emissions inventories from other countries show that COPERT model is commonly used among parties to estimate emissions from road transportation.

Data flow is summarized in [Figure 3](#) from where several main steps may be identified as follows:

- Estimate kilometres driven per vehicle type, driving conditions and vehicle technology;
- Estimate fuel consumption per vehicle type, driving conditions and vehicle technology (bottom-up approach);
- Correct fuel consumption using bottom-up approach in conjunction with top-down approach;
- Emission factors, corrected for hot and cold-start emissions;
- Determine emissions from kilometres driven or fuel consumption

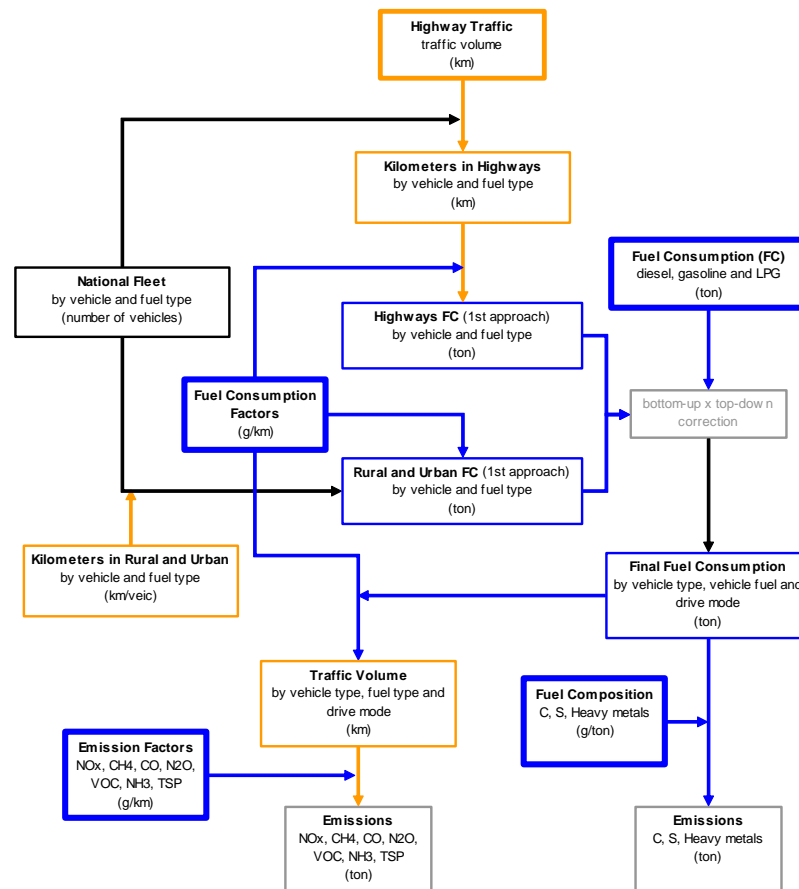


Figure 3. Methodology flow chart for road transport emission calculation.

The number of vehicles in active fleet was derived from sales and abatements statistics and adapted to EMEP/CORINAIR classification.

Table 1. Vehicle fleet.

Vehicle Type		1990	1995	2000	2003
Light Vehicles	Passenger Cars	1,549,449	2,421,077	3,221,482	3,456,494
	Light Duty Vehicles	478,615	710,583	1,045,933	1,163,035
	Mopeds	834,692	708,016	581,340	505,335
	Motorcycles	51,394	111,836	175,957	200,735
	Sum of Light Vehicles	2,914,149	3,951,512	5,024,712	5,325,599
	% of Light Vehicles	96.4	96.6	97.0	96.9
Heavy Vehicles	Heavy Duty Vehicles	103,000	134,000	149,000	162,000
	Bus	1,132	1,355	1,482	1,616
	Coach	5,433	5,249	5,799	4,376
	Sum of Heavy Vehicles	109,565	140,604	156,281	167,992
	% of Heavy Vehicles	3.6	3.4	3.0	3.1
	TOTAL VEHICLES	3,023,714	4,092,116	5,180,993	5,493,591

Vehicle exhaust emissions and fuel consumption are strongly dependent on speed. Three driving modes were individualized in accordance with source categories SNAP97 from CORINAIR/EMEP methodology: urban, rural and highway. For each driving mode average speeds had to be set by vehicle type. Several information sources were used to establish average circulation speeds for each vehicle type.

Table 2. Assumed vehicle speeds by driving mode and vehicle type.

Driving Mode	Vehicle Type	Assumed Speed (km/h)	Source
Highway	Passenger Car (PassCar)	124	PNAC, 2003
	Light Duty Vehicles (LDV)	124	PNAC, 2003
	Heavy Duty Vehicles (HDV)	90	Maximum Legal Value
	Coaches	90	Maximum Legal Value
	Motorcycles	124	PNAC, 2003
Rural	Passenger Car (PassCar)	70	Maximum Legal Value
	Light Duty Vehicles (LDV)	70	Maximum Legal Value
	Heavy Duty Vehicles (HDV)	60	Maximum Legal Value
	Coaches	60	Maximum Legal Value
	Mopeds	40	Maximum Legal Value
	Motorcycles	70	Maximum Legal Value
Urban	Passenger Car (PassCar)	24.9	Gois et al., 2005
	Light Duty Vehicles (LDV)	24.9	Gois et al., 2005
	Heavy Duty Vehicles (HDV)	24.9	Gois et al., 2005
	Buses	14.8	Carris, 2005
	Coaches	24.9	Gois et al., 2005
	Mopeds	24.9	Gois et al., 2005
	Motorcycles	24.9	Gois et al., 2005

Fuel consumption under highway driving mode was estimated for each fuel using a bottom up approach based on estimates of distance driven in this driving mode.

$$Highway_{FC(f,y)} = \sum_c \sum_t [Highway_{Km(c,t,f,y)} \times FC_{(c,t,f,Hway)}] \times 10^{-6} \quad (1)$$

Where,

- $Highway_{FC(f,y)}$ = fuel consumption of fuel type f in highway driving mode by vehicles of all classes in year y (km/yr);
- $Highway_{km(c,t,f,y)}$ = total kilometres driven in highway net-road by vehicles of class c, with technology t, using fuel f in year y (km/yr);
- $FC_{(c,t,f,Hway)}$ = fuel consumption factor for vehicle type c, with technology t, using fuel f in highway driving mode (g/km);
- c = vehicle class or type: light passenger, LDV, HDV, etc;
- t = vehicle technology: PRE-ECE, ECE, Euro I, Euro II, etc;
- f = fuel type (gasoline, diesel or LPG);
- y = civil year.

Fuel consumption under urban and rural driving modes was estimated simply by subtracting fuel consumption estimated in highway mode from total fuel sales, at national level.

Individual fuel use under rural and urban driving conditions was finally determined from the number of vehicles, kilometres driven in urban and rural modes and, fuel consumption factors for all vehicle categories according with equation 3.

$$\begin{aligned} Rural_{1stFC(c,t,f,y)} &= T_{class(c,t,f,y)} \times Km_{rural(c,f,y)} \times FC_{(c,t,f,s)} \times 10^6 \\ Urban_{1stFC(c,t,f,y)} &= T_{class(c,t,f,y)} \times Km_{urban(c,f,y)} \times FC_{(c,t,f,s)} \times 10^6 \end{aligned} \quad (3)$$

Where,

- $Rural_{1stFC(c,t,f,y)}$, $Urban_{1stFC(c,t,f,y)}$ = first approach fuel consumption in rural and urban areas made by vehicles of class c, with technology t, using fuel f in year y (t);
- $T_{class(c,t,f,y)}$ = number of vehicles of class c, with technology t, using fuel f in year y;
- $Km_{rural(c,f,y)}$, $Km_{urban(c,f,y)}$ = rural and urban kilometres driven per vehicle of class c, using fuel f in year y (km/vehicle);
- $FC_{(c,t,f,s)}$ = fuel consumption factor for vehicles of class c, with technology t, using fuel f, at speed s (g/km).

Fuel adjustments are necessary because presently in the process of splitting total fuel among car types and fuel types not all class percentages add to unity. Therefore a correction must be made to make total fuel consumption equal original statistical

data. Urban and rural fuel consumption estimates were corrected by the following factor for car type c , technology t , fuel f , driving mode d and year y .

$$Correc_{Factor(f,y)} = \frac{[Total_{FC(f,y)} - Highway_{FC(f,y)}]}{\sum_c \sum_t [Rural_{1stFC(c,t,f,y)} + Urban_{1stFC(c,t,f,y)}]} \quad (4)$$

Correction factors are later applied to the first approach fuel consumption under rural and urban driving conditions in the following manner:

$$Urban_{FC(f,y)} = Correc_{Factor(f,y)} \times \sum_c \sum_t [Urban_{1stFC(c,t,f,y)}]$$

$$Rural_{FC(f,y)} = Correc_{Factor(f,y)} \times \sum_c \sum_t [Rural_{1stFC(c,t,f,y)}] \quad (5)$$

This correction guarantees that emission estimates are in accordance with good practices (IPCC, 2000; IPCC, 1996). Although emissions were derived from estimate of vehicle driven kilometres and from fuel consumption per kilometre (bottom-up approach), they were corrected for total national fuel sales (top-down correction).

Final activity, in kilometres, is estimated according with total fuel consumption in the following manner:

$$Km_{urban(c,t,f,y)} = \frac{[Urban_{FC(c,t,f,y)} \times 10^6]}{FC_{(c,t,f,s)}}$$

$$Km_{rural(c,t,f,y)} = \frac{[Rural_{FC(c,t,f,y)} \times 10^6]}{FC_{(c,t,f,s)}} \quad (6)$$

$$Km_{total(c,t,f,y)} = Km_{highway(c,t,f,y)} + Km_{urban(c,t,f,y)} + Km_{rural(c,t,f,y)}$$

Where,

- $Km_{total(c,t,f,y)}$, $Km_{highway(c,t,f,y)}$, $Km_{urban(c,t,f,y)}$, $Km_{rural(c,t,f,y)}$ = total driven distance under all driving modes estimated for vehicles of class c , with technology t , using fuel f in year y (km);
- $Urban_{FC(c,t,f,y)}$, $Rural_{FC(c,t,f,y)}$ = total fuel consumption allocated to vehicles of class c , equipped with technology t , using fuel type f , under urban and rural driving conditions in year y (t);
- $FC_{(c,t,f,s)}$ = fuel consumption factor for vehicles of class c , with technology t , using fuel f , at speed s (g/km).

3. RESULTS

Distance driven has increased steadily between 1990 and 2002. The increase in highway circulation, which has grown 7.8 times in twelve years, reflects not only the growth of the Portuguese highway road-net, but also an increase in intensity of road use. For the same period, rural and urban circulation has increased 2.0 and 1.6 times, respectively. Total road traffic activity has increased 110.7% since 1990. From 2002

to 2003 total road traffic activity registered a slight decrease of 0.47%, probably the result of an increase of oil prices in last years.

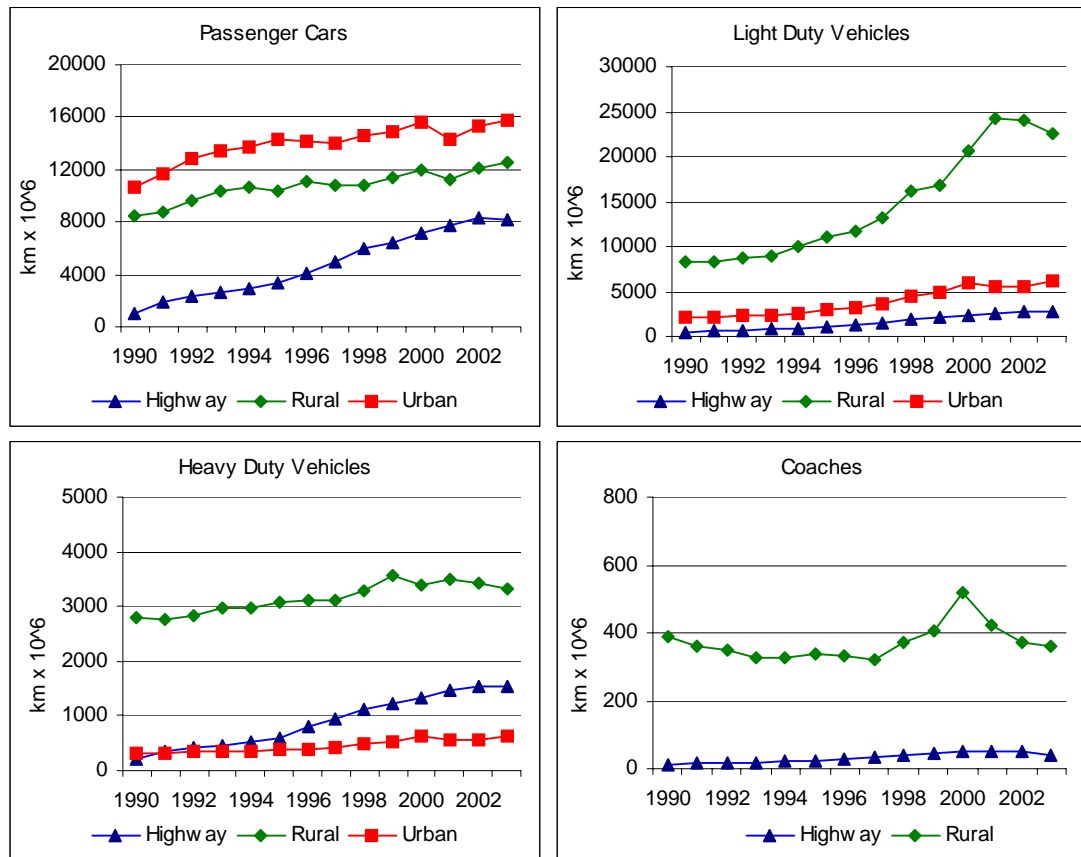


Figure 4. Distances travelled.

Exhaust greenhouse gases (GHG) emissions from road transportation were estimated about 19.3 Mt CO₂ equivalent in 2003 representing an increase of 104.8% compared to 1990 emission level estimated about 9.4 Mt CO₂ equivalent (Figure 5). Between 2002 and 2003 GHG emissions registered a slight decrease from 19.4 to 19.3 Mt CO₂ equivalent. Emissions of N₂O have increased by a factor of 4.2 since 1990 due to the introduction of catalytic converters. Some authors suggested that in some cases N₂O emissions could increase by as much as a factor of 10 (Wade et al., 1994; de Soete and Sharp, 1991; Dasch, 1992). The introduction of catalytic converters brought also some disadvantages including the increase of CO₂ and NH₃ emissions which contribute to climate change and acid deposition. It is difficult to assess the extent to which CO₂ emissions have increased as a result of fitting catalytic converters, because improvements in fuel economy have been made at the same time as development of the engine management systems that are required to minimise NO_x and VOC emissions.

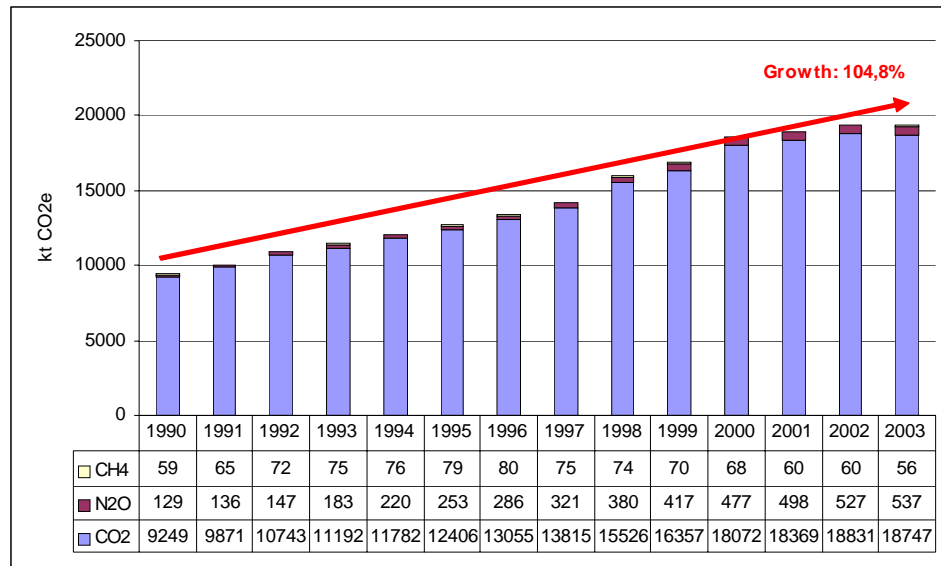


Figure 5. Estimated emissions from road transportation.

Since 1992 that catalytic converters had contributed for major reductions of pollutants with special concern to urban air quality such as CO, NO_x, VOC and NMVOC (see Figure 6). Although NO_x absolute emissions has increased 22.7% since 1990 the emission factor given in mass of pollutant by fuel unit decreased from 14.9 kgNO_x/tep in 1990 to 4.6 kgNO_x/tep in 2003.

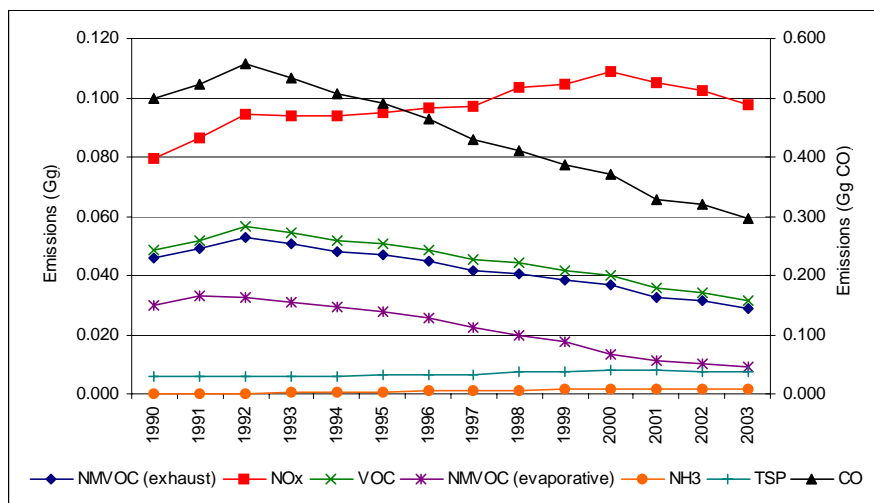


Figure 6. Non greenhouse gases emissions.

The model performance was evaluated by comparing the estimated fuel consumption (first approach bottom-up before top-down correction) with the real fuel consumption. Estimated fuel consumption varies within a maximum of $\pm 30\%$ from real fuel consumption for all period of analysis. Very good estimations were performed for 1997 where gasoline and diesel differs from real values only by -2% and 0.2% , respectively.

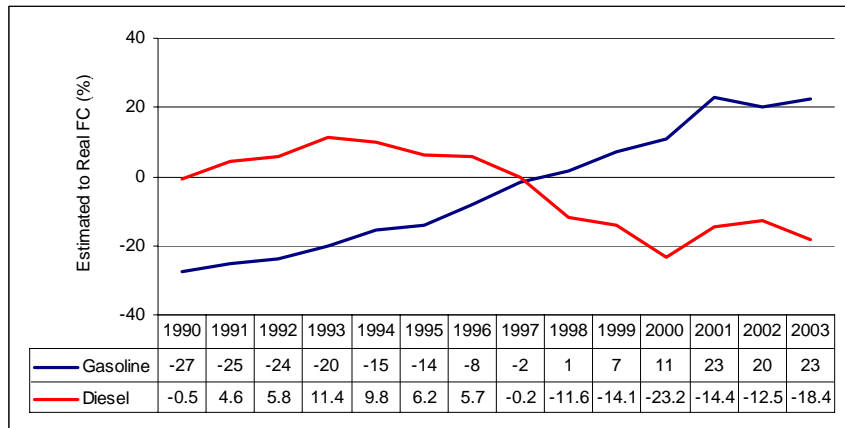


Figure 7. Estimated to real fuel consumption.

The high level of disaggregation used in this inventory could be very useful to quantify emissions changes resulting from specific measures. It could be interesting to evaluate the impact of speed changes for each vehicle category and driving mode. The increase of average speed in urban areas could lead to a decrease in emissions. Obviously, this increase in average speed should be achieved by decreasing traffic jams instead of increasing the speed limit in urban areas. In highways, the decrease of the average speed could contribute to significant emission reductions particularly for light vehicles.

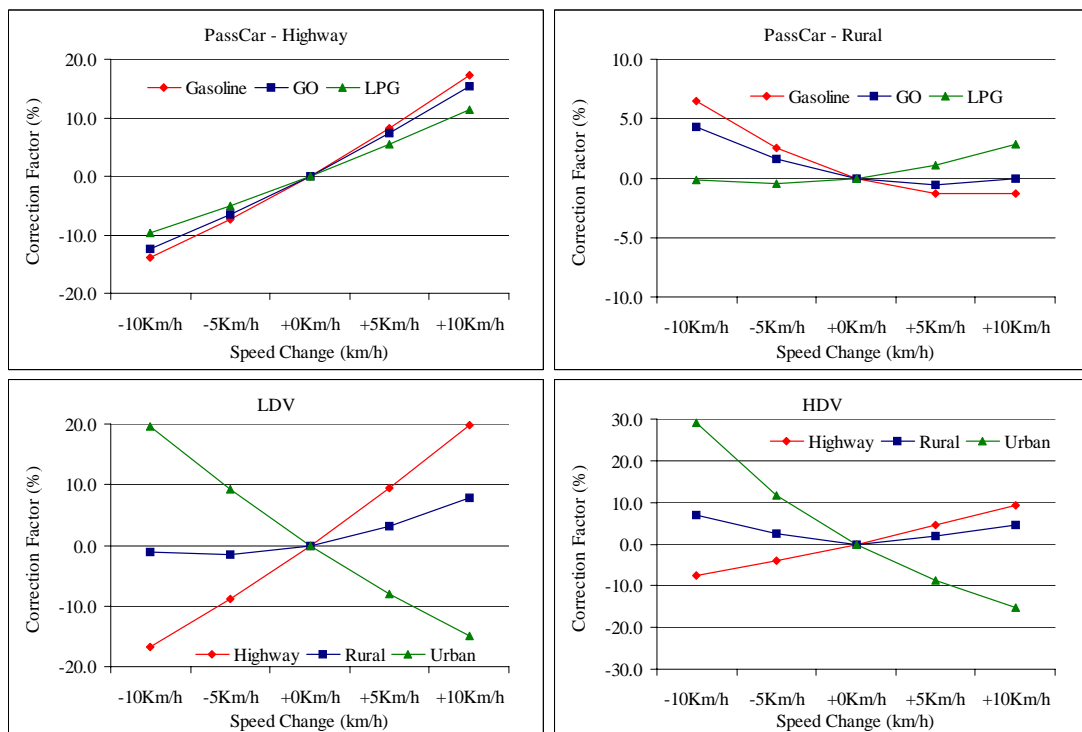


Figure 8. Emission correction as function of speed.

4. CONCLUSION

The use of the bottom up approach to estimate national emissions from road transportation should be considered a good approach given the similarity between the real and predicted total fuel consumption. Important results were achieved when considering the level of emission disaggregation within this sector.

However some problems were identified when scaling down the level of disaggregation in road transportation. This sector is one of the most complex sectors when overall emissions need to be calculated. The amount of variables and the data required for the calculation is scattered amongst several institutions and sometimes is simply inexistent. Therefore, many assumptions must be made increasing the model uncertainty. No specific calculations were performed to assess the model uncertainty however this subject is crucial to support the model output results.

Further work is needed to improve model accuracy and some topics for research were identified during the present study: use of a vehicle mileage correction factor; estimate emissions of resuspended particulate matter from tyre and brake wear; use the road gradient correction factor; improve temporal allocation for line and are sources; apply quality assurance and quality control procedures; estimate model uncertainty. The model accuracy could still be improved by deriving the vehicle activity variable from the data acquired at the vehicle inspection centres which are under the authority of the *Direcção Geral de Viação*.

5. ACKNOWLEDGEMENTS

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STUDY OF MTBE IN TEHRAN'S ATMOSPHERE

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ABSTRACT

Around 3-million motor vehicle in Tehran burn above 20-million of liters gasoline daily. This is the first time that MTBE (Methyl tert-butyl ether) levels in Tehran have been extensively characterized.

In order to evaluate the levels of MTBE pollutant in the Tehran, ambient air samples were collected in sorbents tubes and analyzed with a gas chromatograph according to procedures described in the NIOSH method.

The aim of this study was determination of MTBE concentration and its role on Tehran pollution. For each collected sample, MTBE were analyzed by using a GC with a flame ionization detector (FID). Quality assurance was performed collecting duplicate samples which were analyzed in replicate to quantify the precision of air quality measurements.

The established calibration curves for the MTBE samples were found with R²-value > 0.995. Mean values of MTBE concentrations in Tehran atmosphere were 0.1- 0.55 ppm.

Key Words: MTBE, Urban air quality, Vehicular emission, Tehran pollution

1.INTRODUCTION

MTBE (Methyl tert-butyl ether) is important organic air contaminants emitted into the atmosphere by automobile exhaust, fuel evaporation, storage, transfer and handling of fuel (Donati, 1995). The greatest health risk from exposure to MTBE is carcinogenic and neurotoxic (Jo and Choi, 1996).

Its poisoning effect may occur due to inhalation , ingestion or fast absorption through the skin (Cocheo, 1998). The main symptoms are : headache , confusion , and loss of the muscle control , irritation of the airways (Wallac, 1996).

Traffic - induced emissions are a major source of urban air pollution (Rogak et al., 1997). As a result of increased emissions and inadequate ventilation , the concentration of these pollutants in confined spaces can result in deleterious impacts on health and the well being of exposed population (Swati, 1999).

Pollutants in urban atmosphere arise from traffic and in particular, are emitted from automotive exhaust or fuel evaporation from tanks, vehicle carburetors, and gasoline stations during the refilling operations (Faiz et al., 1996). To date, the traffic density

of the transportation system has increased dramatically during the recent years in many developing and developed countries (Meek, 2000 and Sonawance et al., 2000). Tehran is amongst a few capitals of the world, which mountains surround the city from the North and East. The total area of the city is 800 km². There are four accurate seasons, with the annual mean rainfall at about 230 mm. The basin's topographical situation does not allow, the major part of the time, the free circulation of winds and good ventilations. A major focus of this study is the contribution of vehicular emissions to air pollution in Tehran. Vehicular traffic can be classified into three categories: gasoline fueled (cars), diesel fueled (buses, trucks) and motorcycles (2-stroke and 4-stroke). In recent years due to consumption gasoline content of MTBE (instead gasoline leaded) in Tehran, evaluation of MTBE in atmosphere were necessary.

It is estimated that 80 – 85 % of MTBE in the atmosphere comes from the automobile depend on traffic flow and street geometries (Rama, 2004). MTBE is the most widely used fuel oxygenate and is added to gasoline at 5-15 % in gasoline. MTBE has accumulated sufficient evidence as a human carcinogen (Johnson et al., 2000 and Squillace et al., 1996).

Evaporative emissions from vehicle systems are generally grouped into three categories namely (Meek, 2000) :

- diurnal emissions
- hot - soak emissions
- running losses

2.EXPERIMENTAL METHOD

The method presently adopted for air sampling is based on adsorption on a activated sorbent tube and desorption methods followed by gas chromatography analyses. The choice of the sampling points in the monitoring campaign followed two basic criteria. First a representative ness of the area, in areas with high population density or intense traffic. Second, in the surroundings of the main sources.

The sampling height was about 1.6 m above the ground. The preparation and pre-conditioning of the instruments performed prior to sampling. MTBE (99.5 %) used were of analytical grade (MERCK CO.)for standard solutions. A calibration curve was prepared from known concentrations of MTBE. Carbon disulfide (analytical grade) used for making all solutions the method 1615 which was suggested by the US National Institute for Occupational Safety and Health (NIOSH) was adopted. In this sampling, sampling system consisted of a sorbet tube (SKC charcoal tube, 100 mg/50 mg) and low-flow sampling pumps (model 224-44EX, SKC Inc.). Air was pumped (2lit/min) through absorption tubes, which contained two sections.

The charcoal sampling tube should always be vertical during sampling to prevent channeling of the charcoal. Generally, the loss of the MTBE has to be minimized during sampling, samples were stored in the dark at 4 °C before analysis no longer

than 24 – 48 hr later. After sampling, the charcoal tube is removed from the sample holder and both ends capped with plastic caps provided. During the sampling procedure one charcoal tube is opened at the sample site and the ends capped .No air is drawn through this tube which serves as a blank.

Desorption of the collected samples from the charcoal tube have been accomplished by CS₂. The analyze of samples were carried out using a gas chromatograph (GC - Perkin Elmer - Sigma 3B) with FID detector. The established calibration curves were found with R²- Value > 0.95. The monitoring was performed in twenty stations in strategic with in the municipal district.

3.RESULTS AND DISCUSSION

Organic compound in Tehran atmosphere originating in automobile engine emissions are among the most significant sources of air pollution in the congested and poorly ventilated streets of the city. It is important to note that the Iran vehicle fleet differs significantly from fleets in other developed countries due to the fact that most of those vehicles have no evaporative emission control devices, even in many of the new vehicle models. Fuel in Iran has been supplied at one of the cheapest prices. The availability of low cost fuel has increased private vehicle ownership. Around 3 million motor vehicle burn above 15-20 million of liters gasoline daily.

In recent years due to consumption gasoline content of MTBE (instead gasoline leaded) in Tehran, evaluation of MTBE in atmosphere were necessary. Table 1 shows the mean concentration of MTBE obtained in the Tehran's atmosphere of the sampling sites.

The finding shows that the mean concentration of MTBE is 337 ppb with a maximum of 1000 ppb.

Table 1-Average concentration of MTBE (ppb)

Sampling sites	MTBE	Sampling sites	MTBE
Azadi seq.	230	Shoosh seq.	200
Enghelab seq.	350	Khorasan seq.	150
Ferdouci seq.	200	Bazar	450
Emam hossein seq.	220	Peroozi str.	1000
Damavand str.	520	Khavaran str.	350
Kauj seq.	10	Nemat-abad str.	150
Vanak seq.	330	Shahr- Ray	300
Azadi str.	200	Elghadeer str.	500
Naser Bridge	200	Shaheed Beheshty str.	700
Tajreesh seq.	350	Rah- Ahan seq.	N.D.

In Iran, air-quality standards have not yet been set for organic pollutants. It is accepted that MTBE is genotoxic carcinogen and therefore no absolutely safe exposure level can be defined.

There is evidence that humans (specially children) who live near heavily traveled streets and squares that has showed in the table may be at an elevated risk of cancer, including leukemia (U.S. EPA, 1986 and U.S. EPA, 1996).

It is known that the fleet age significantly affect the exhaust of toxic contents from a vehicle. Figure 1 shows daily concentration of MTBE in Tehran.

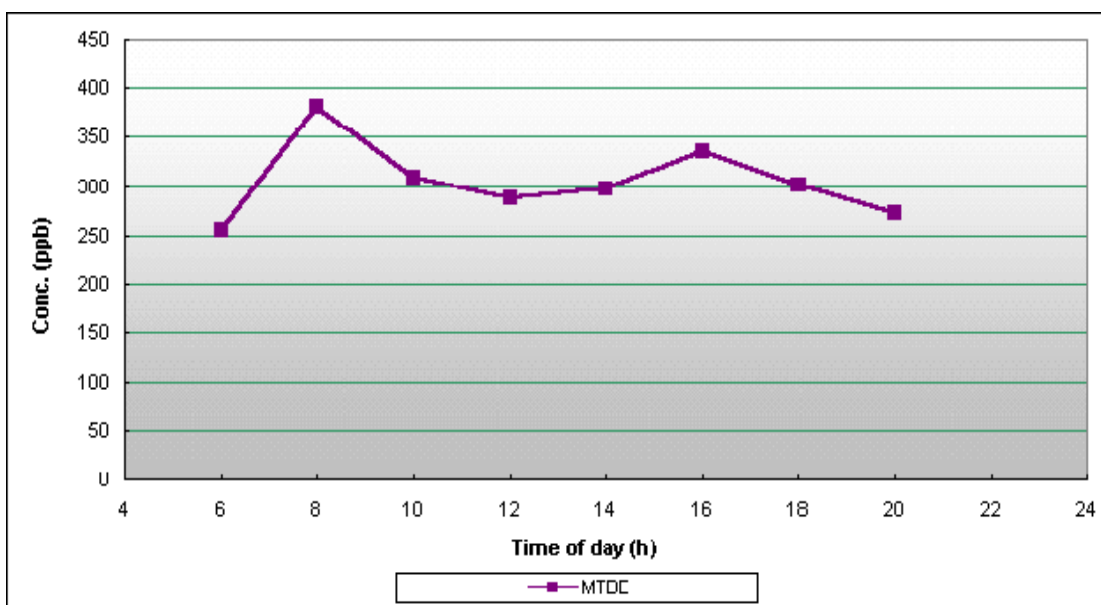


Figure 1 : Fluctuation of daily concentration of MTBE

In addition the area of two gas station and surrounding have been selected for sampling MTBE (Table 2-3). These stations are influenced directly by MTBE.

Table 2- Contents of average of MTBE in the different distance of Khavarun Gas station during the summer and winter seasons.

Compound	Jun. and Jul.	Dec. and Jan.
Sampling site	(ppm)	(ppm)
Gas Station area	5.85	5.4
8m from Gas station	1.1	1.1
25m from Gas station	0.45	0.25
50m from Gas station	0.13	0.18
100m from Gas station	0.45	0.12

Because MTBE is highly volatile because of its high vapor pressure (245 - 276 mm Hg at 25 °C) therefore the concentration of this compound is become more in the summer time (IAOF, 1997).

Table 3 - Contents of average of MTBE in the different distance of Beheshtee Gas station during the summer and winter seasons.

Compound Sampling site	Jun. And Jul. (ppm)	Dec. and Jan. (ppm)
Gas Station area	3.4	3.1
8m from Gas station	1.1	2.6
25m from Gas station	0.68	1.5
50m from Gas station	0.55	0.35
100m from Gas station	0.48	0.22

Environmental exposure to MTBE can be largely lowered by adopting preventive measures including traffic restrictions, equipped of new cars with catalytic converters and reformulation of gasoline and devices to recover vapors from gasoline pumps. There is evidence that humans (specially children) who live near heavily traveled streets and squares that has showed in the table may be at an elevated risk of cancer, including leukemia. Benzene is a mutagen and human teratogen.

No safe guide lines for airborne MTBE can be recommended as they are carcinogenic to humans and there are no known safe threshold level. Legislation should also be introduced and enforced to promote the use of reformulated gasoline with low MTBE content.

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AIR QUALITY NEARBY MOTORWAYS: DEVELOPING NEW MONITORING METHODOLOGIES

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ABSTRACT

A study has been carried out to evaluate the air quality impact of the motorway ‘A2 South’ that links Lisbon to Faro in Portugal. The monitoring of atmospheric pollutants with the use of two simultaneous measurement methods (indicative and continuous measurements) was one of the strategies to evaluate the conformity with the air quality legislation. This paper gathers the results obtained in two air quality monitoring campaigns. Four sampling points located along three sections were selected. Several pollutants were measured under different sampling methodologies. Meteorological and traffic data were also collected. High level concentrations of particles were found close to the motorway toll booths, and benzene seems to be the best descriptor to evaluate the impact on air pollution.

Key Words: Motorway air emissions; air quality monitoring; indicative and continuous measurements; traffic measurements; pollutants dispersion.

1. INTRODUCTION

Air pollution represents one of the biggest threats to public health and ecosystems. The fast technological progress of the modern world brought an increase in the amount of pollutants deposited in the atmosphere, harming in a very serious way the quality life of our planet (Castro *et al.*, 2003). The pollutants emitted by traffic and the consequent effects in public health are assuming a larger relevance. That has been verified by epidemiological studies developed in different capitals and cities in order to establish relationship patterns with air quality (Castro *et al.*, 2003).

The transport sector is also one of the most important sources of greenhouse gases like carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). It is also responsible by the emission of pollutants associated with transboundary transport such as sulphur oxides (SO_x), nitric oxides (NO_x), carbon monoxide (CO), and nonmethane volatile organic compounds (COVNM) (Borrego *et al.* 2002). This activity also brings substantial emissions of particles and heavy metals (Eggleston *et al.*, 1989).

The turbulence induced by traffic circulation is a determinant factor in the dispersion of pollutants in the vicinity of roadways (Rao *et al.*, 2002). Vehicles contribute for

the degradation of air quality, essentially through the emission of CO, hydrocarbons (HC), NO_x and particles (Mihau and Boulhol, 1990). Once introduced in the atmosphere, pollutants are transported by the wind and diluted under a dispersion process. In many cases, a chemical transformation or deposition can also occur. Monitoring emissions from line sources has increased in the last years in order to make possible to establish relations between the deterioration of air quality and the increment in the number of vehicles (Hansel *et al.*, 2004).

Pollutants concentrations depend on many parameters and change considerably as a function of the atmospheric conditions, mainly wind. In roadways, pollutants are randomly emitted since traffic volume and speed are dependent on the human behaviour. Thus, these two practically independent factors define the environmental impact of the roadway (Murgel *et al.*, 1997). However, the methodology of continuous measurements is associated with a very high cost and a constant operational activity and logistic, only promoting the control of the area where the monitoring station is installed (Murgel *et al.*, 1997).

The air quality in Portugal is known mainly from the data collected in monitoring stations predominantly located in the great urban areas (areas of traffic influence), and in some industrial areas, but also through multiple environmental impact assessment studies (Ferreira *et al.*, 2001). Moreover, numerical air quality modelling is contributing to the evaluation of the national air pollutants levels and distribution (Monteiro *et al.*, 2005).

The A2 *South* motorway is characterized for being one of the first existing projects in Portugal within the new environmental impact evaluation regulations where assessment goes behind construction. The objective of this study was to evaluate the traffic impact in the air quality in three sections of the “A2 South”.

2. THE CASE-STUDY OF A2 SOUTH MOTORWAY: AIR QUALITY EVALUATION METHODOLOGY

Three main sections of the A2 South (sections Castro Verde / Almodôvar, Almodôvar / S. Bartolomeu de Messines and S. Bartolomeu de Messines / Via Longitudinal do Algarve - VLA) were studied using two measurement-monitoring methodologies. Four sampling locations were selected: Castro Verde, Gomes Aires, Paderne, and VLA. The campaigns took place between March 8 and April 12, 2004 (1st campaign), and between June 28 and July 26, 2004 (2nd campaign). Nitrogen dioxide, ozone (O₃), SO₂, particles, CO, and benzene (C₆H₆) were measured under different sampling methodologies.

The studied roadway has a peculiar characteristic meant for intense traffic peaks associated with the seasonal affluence increase since it bridges a connection to a very required tourist area (Algarve Region). Daily mean traffic of about 7 000 vehicles, and in peak days around 26 000 vehicles, were identified during the campaigns.

The campaigns implied the continuous measurement of gases and particles, and the weekly averaged concentrations of SO₂, NO₂, O₃ and C₆H₆ with diffusion sampling tubes placed at some particular points, previously chosen. Also, meteorological parameters were simultaneous measured due to their influence in the dispersion of atmospheric pollutants, namely wind speed and direction, rainfall, temperature and humidity. The meteorological station was always placed in the same location where pollutants were monitored - in the vicinity of the SNIF - AIRLAB (mobile laboratory of the Universidade Nova de Lisboa - Figure 1), and close to the places where the indicative measurement with diffusion tubes was taking place.



Figure 1: Aspects of the interior and exterior of the mobile laboratory - SNIF AIRLAB and its measurement equipment

The rank of diffusion tubes at different distances of the berm and at both sides of the roadway, aimed to provide a better understanding of the dispersion pattern of the pollutants along those areas (Figure 2).

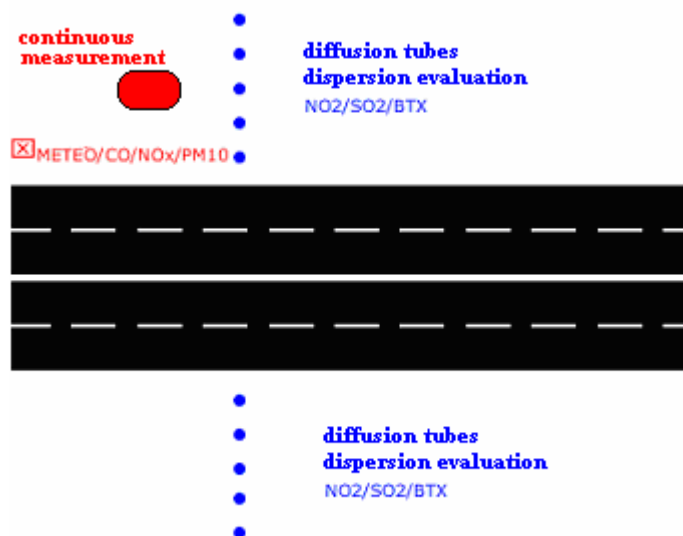


Figure 2: Project of installation of the measuring equipment and SNIF Lab.

2.1 Gases Sampling

The monitoring of pollutant gases was made using passive Radiello® diffusion tubes for the sampling of SO₂, NO₂, BTX (benzene, toluene, and xylene) and O₃. Toluene and xylene are pollutants not currently legislated in terms of air quality. This equipment allows the collection of an integrated pollutant sample for one determined period of exposure. The result corresponds to the average concentration for the chosen period of exposure. The exposure time of the diffusers in the sampling points was one week. Diffusion tubes were used in Paderne (close a major toll booth), and Gomes Aires.

CO and NO₂ were measured using a chemiluminescence continuous measurement technique with integrated average concentrations of fifteen minutes, which were compiled in hourly average values. For CO, taking into account the legislated eight-hours base averaged value, this integrated period was used to allow a comparison with the air quality limit value foreseen in the Decree-law n.º 111/2002 of April 16.

2.2 Particle Sampling

The monitoring of particles was carried through a continuous beta radiation instrument. The Decree-law n.º 111/2002, in its Annex XI (Section IV), indicates that the method of reference for sampling and measuring of particles PM₁₀ is the one described in the European norm EN 12341. This norm considers the gravimetric method as the reference method. However, this legislation allows the use of other methods if demonstrated that their results are equivalent to the ones of the reference method. Therefore, a correction factor of 1.18 to the PM₁₀ collected data (correction factor for traffic stations) was applied on the basis of inter-comparison studies performed by the Portuguese Institute of the Environment. Average concentrations of fifteen minutes were collected and integrated as hourly average values. Since the legislation establishes daily and annual limit values, the hourly data were then integrated on a daily basis to aim the respective comparison with the Decree-law n.º 111/2002 of April 16.

2.3 Meteorology

The meteorological conditions influence the concentration and the distribution of the atmospheric pollutants; therefore, the measurement of the meteorological parameters was performed in all sampled locations. A portable meteorological station measuring wind direction and speed, rainfall, temperature, and sun radiation, was used for this purpose.

3. RESULTS FROM CONTINUOUS AND INDICATIVE MEASUREMENTS

The results obtained were compared with the current legislation. The comparison was made with the legislative values for the year 2005 (Limit value - LV and/or Limit value + Margin of Tolerance - LV + MT), assuming different time bases (hourly, octo-hourly, daily or annual), depending on the pollutant. Measured data were compared with the designated values (in bold) in Table 1.

Table 1: Limit-values or Limit-values + Margin of Tolerance used for comparison of results

polutant	temporal base	limit value ($\mu\text{g}/\text{m}^3$)	tolerance value 2005	final value 2005
PM ₁₀	$\mu\text{g}/\text{m}^3$ (Diário)	50 $\mu\text{g}/\text{m}^3$ ^{**}	-	50 $\mu\text{g}/\text{m}^3$
PM ₁₀	$\mu\text{g}/\text{m}^3$ (Anual)	40 $\mu\text{g}/\text{m}^3$ ^{**}	-	40 $\mu\text{g}/\text{m}^3$
NO ₂	$\mu\text{g}/\text{m}^3$ (Horária)	200 $\mu\text{g}/\text{m}^3$ ^{**#}	50 $\mu\text{g}/\text{m}^3$	250 $\mu\text{g}/\text{m}^3$
NO ₂	$\mu\text{g}/\text{m}^3$ (anual)	40 $\mu\text{g}/\text{m}^3$ ^{**#}	10 $\mu\text{g}/\text{m}^3$	50 $\mu\text{g}/\text{m}^3$
CO	mg/m ³ (Octo-horária)	10 mg/m ³	-	10 mg/m ³
SO ₂	$\mu\text{g}/\text{m}^3$ (Diária)	125 $\mu\text{g}/\text{m}^3$ ^{**}	-	125 $\mu\text{g}/\text{m}^3$
C ₆ H ₆	$\mu\text{g}/\text{m}^3$ (Semanal)	5 $\mu\text{g}/\text{m}^3$ ^{**#}	5 $\mu\text{g}/\text{m}^3$	10 $\mu\text{g}/\text{m}^3$

^{*}2005

^{**}2010

Figures 3 and 4 present the measured PM₁₀ values in Paderne and Castro Verde locations.

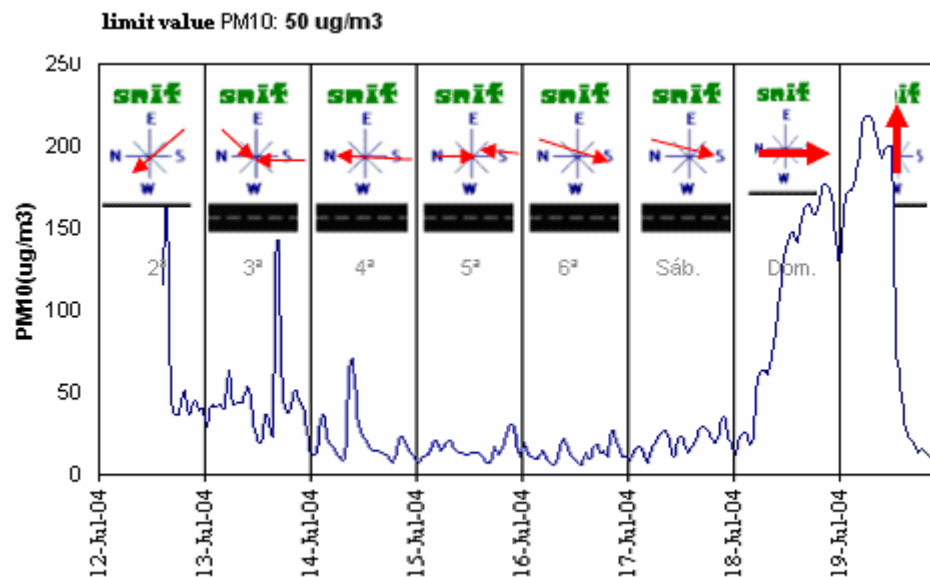


Figure 3: PM₁₀ concentrations - Paderne (2nd campaign)

It was verified that PM₁₀ concentrations measured during the 2nd campaign rose significantly during a weekend period. In order to evaluate the results, PM₁₀ data from the two closest background monitoring stations were used. A natural event due to the wind transport of particles from the Sahara desert was identified as the cause for such high levels.

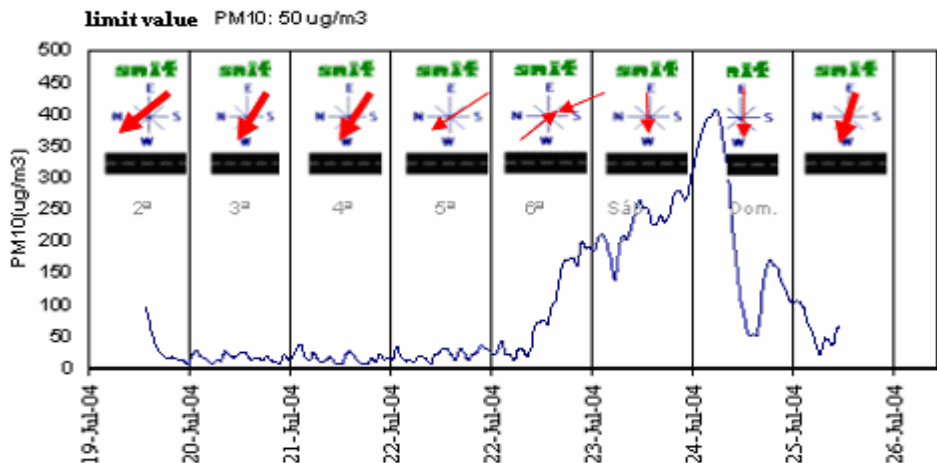


Figure 4: PM₁₀ concentrations - Castro Verde (2nd campaign)

The air quality monitoring stations chosen to make the comparison were Pontal (background urban station - Portimão) and Chamusca (background rural station - Chamusca). Figure 5 presents the hourly-averaged PM₁₀ values for these three monitoring sites.

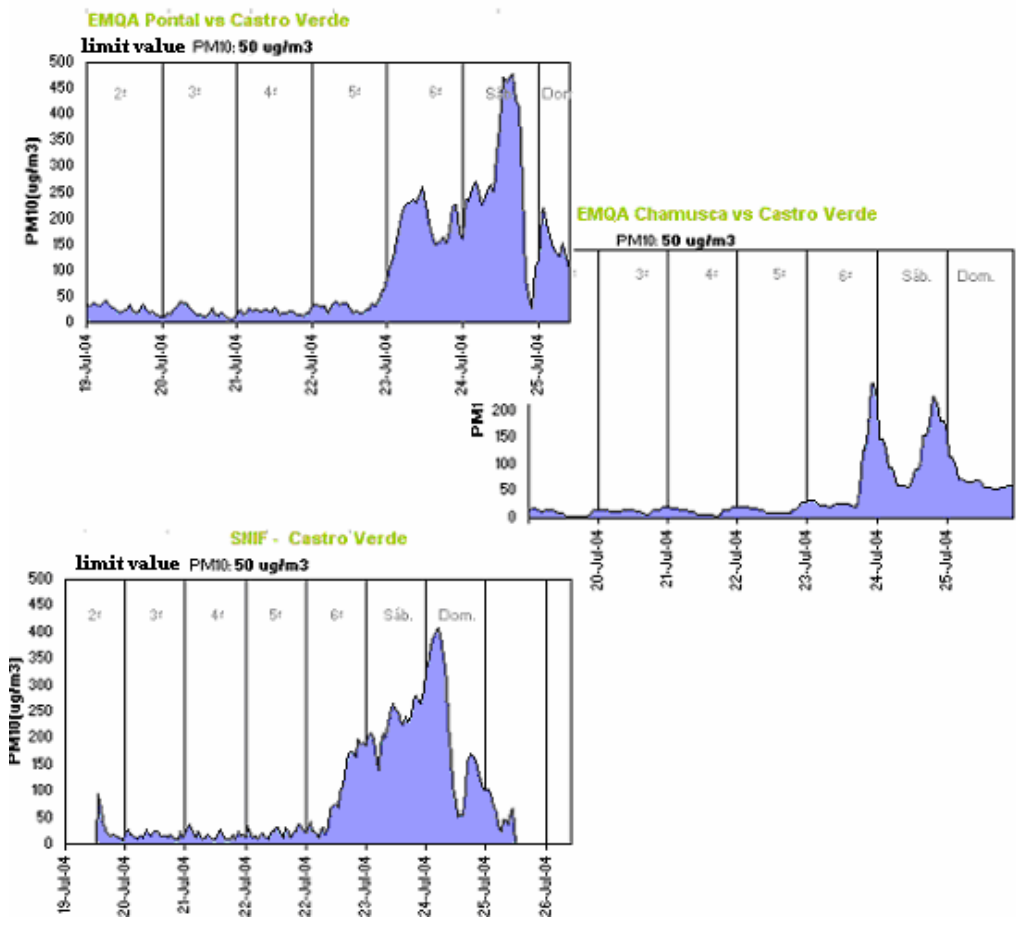


Figure 5: Hourly averaged concentrations of PM₁₀ in the Pontal, Chamusca and Castro Verde monitoring sites (2nd campaign)

By the analysis of the data, it can be concluded that the high concentrations during the sampling period (through the AIRLAB) were mainly due to a particles event, given the similar levels and trends found in these monitoring stations (Figure 5).

However, by the analysis of traffic data during the peak dates, traffic hourly average values (TMH) were very high (maximum TMH of 2290 vehicles - Saturday) and that should have also contributed to the higher particle's level.

The C_6H_6 weekly averaged concentrations, in both campaigns, can be observed in Figure 6.

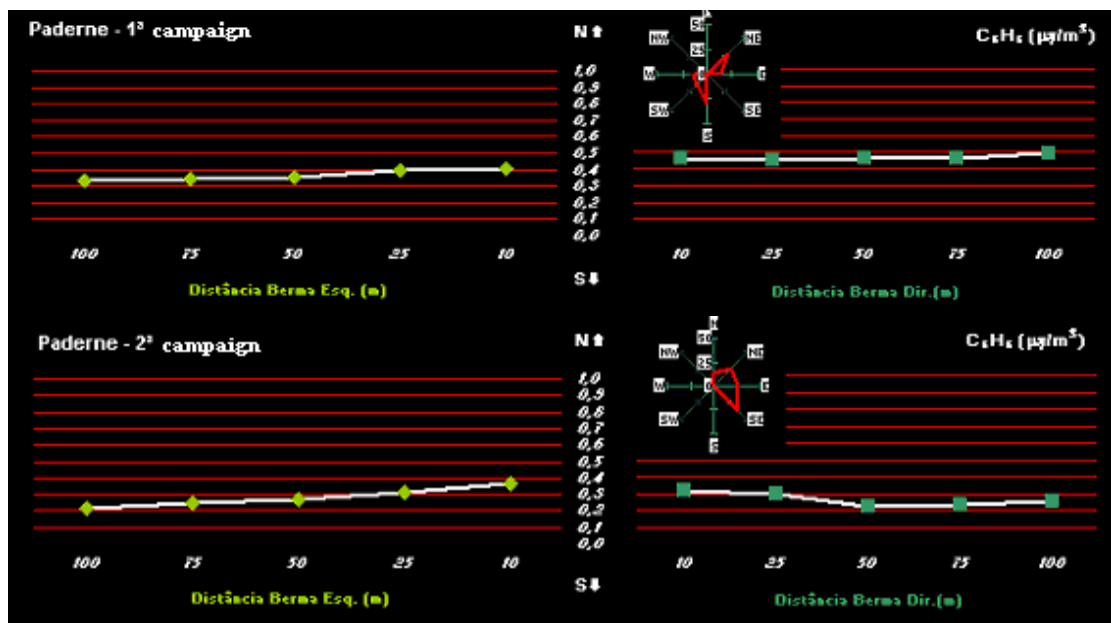


Figure 6: C_6H_6 weekly averaged concentrations in Paderne.

The values collected for C_6H_6 remained low and are similar for the two campaigns, although slightly higher in the 1st campaign. Tendentially, a decreasing pattern is verified from the road to both sides, with exception of one point. A direct comparison with the legislation is not possible because benzene concentrations correspond to weekly averages and not annual averages.

4. DISCUSSION

In general, the values obtained along the two campaigns for all the pollutants were low. For the SO_2 , NO_2 , O_3 , and C_6H_6 gases referring to weekly averages (through indicative measurement - diffusion tubes), it was confirmed that during the accomplishment of the two campaigns in the four monitored points, the measured concentrations were very low. The NO_2 and CO continuous monitoring also registered low values of concentration.

Relatively to the obtained results through the diffusion tubes, for NO_2 and SO_2 , it was not possible to obtain a linear pollutant dispersion behaviour with the distance

to the road. The reduced measured concentrations of pollutants and the wind variable conditions can explain this not linear trend.

The C₆H₆ values presented a decreasing linear dispersion behaviour and larger expression until 50m distance from the road.

By the application of different methodologies involving a detailed analysis of the evolution of the pollutants concentration and their relationship with background stations located in other locations of the region, two critical points (Paderne and Castro Verde) were identified with higher levels of PM₁₀. These occurrences were directly related with the increase of the vehicles traffic circulating in those sections and with a natural event that brought Sahara dust to the monitored area. Meteorological conditions marked by major occurrences of calm situations during the respective sampling periods also determined the levels found.

Results gave evidence that diffusive samplers may overestimate the concentration values when compared with continuous measurement. This occurs when very low concentrations are measured. Knowledge about the traffic counts for the campaign periods allowed a better explanation of the motorway impact in terms of air quality, through the relationship established with certain observed peaks.

For particles, an analysis of the ratio between the number of daily exceedances of the limit-value and the number of sampling days during both campaigns, allowed to extrapolate that the 35-exceedance days permitted annually would be probably overtaken.

Regarding the measurements of CO in the four points of sampling and for both campaigns, very low concentrations values, in 0.2 mg.m⁻³ order (octo-hourly secular base), were observed, with little significant variations in the different cited points. The behavior of this pollutant to the level of its secular distribution, presents in general, a common profile in relation to the increase of its values during the period of weekend, in particular, Friday and Sunday night, probably because of the addition of traffic typically associated to these days of the week in this roadway.

In relation to NO₂, very low values of concentration had also been observed, with a maximum of 18.0 µg.m⁻³, as an average of the measured concentrations in the point of the VLA (continuous monitoring), quite below the threshold value for the protection of human health (250 µg.m⁻³ - hourly base). Concerning the weekly averages of this pollutant (passive sampling monitoring), measured values in general were higher than those acquired through the continuous measurement. When compared with the respective legislated annual value base (50 µg.m⁻³ for year 2005) all values are below the legislated targets.

The highest weekly average concentration of SO₂ occurred at Castro Verde, reaching 9.1 µg.m⁻³, which should be considered as an outlier when compared with the other collected data, but still well below the daily limit-value of 125 µg.m⁻³. It was mentioned that the variations in the concentrations between sampling campaigns are not significant. However, distinct orders of magnitude are observed considering the

two sampling locations (Paderne and Castro Verde). The concentrations of SO₂ in the Paderne point are similar to the background concentrations existing in the region.

In respect to ozone, this pollutant was measured only in places 100 m far from the road (in locations Paderne and Castro Verde). The existence of this pollutant near the roadway is not directly associated with the traffic road circulation: its formation is dependent on other factors such as the solar radiation or the existence of precursor pollutants that may have distinct sources. Therefore, it is probable that the concentrations registered are in fact background concentrations characteristic of this region for that time of the year.

5. CONCLUSION

The monitoring air quality data of the *A2 South*, only indicates potentials problems of particulate atmospheric pollution, because the high level concentrations measured close to the motorway toll booths that should be associated, more than to the PM₁₀ direct emission through the vehicles exaustion, to the resuspension related with the vehicles circulation and to the occurrence of natural events, namely the long distance transport of particles from North of Africa. Benzene seems to be the best descriptor to evaluate the impact on air pollution.

6. ACKNOWLEDGMENTS

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EMISSION FACTORS OF NON-METHANE HYDROCARBONS FOR SOME WIDELY USED PASSENGER CARS IN TURKEY

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ABSTRACT

The objectives of this study are to measure the non-methane volatile organic carbon (NMVOC's) emissions from passenger cars in Turkey having gasoline engines, to determine emission factors of these vehicles for BTEX compounds and comparison of emission factors obtained in this study with the emission factors of the other countries.

This study was conducted in two parts: The first part was to determine the categories of passenger cars widely used in Turkey, and also to determine the average carbon monoxide (CO) and hydrocarbon (HC) emissions at idle condition for these car types based on the exhaust emission measurements of Ankara Çevre Koruma Vakfı (ANÇEVA). The second part of the study was to analyze the gas composition of exhaust gasses at different road conditions for BTEX components by using gas chromatography.

The highest emission factors among hydrocarbons investigated in this study were found for toluene and m-xylene. Generally, as driving speed increases the emissions of HC's are found to decrease in concentration. It was interesting to note that the highest emissions occur at 30 km/hr speed which is the mostly used speed in crowded streets and busy intersections. Therefore, it was concluded that it is very important to take measures for emissions in the city traffic. Cold start emissions were also found to be higher than the hot start emissions.

Key Words: Emission Factors, BTEX Emissions, CO/HC Emissions, Turkish Fleet of Passenger Cars.

1. INTRODUCTION

1.1. General

The rapid improvement in automotive industry, increase in the life standard, and fast increase in the number of the motor vehicles with increase in population caused air pollution by motor vehicles to become an important problem in the world. Today most of the big cities suffer from air pollution due to traffic.

Air pollution from the road traffic depends on both vehicle emissions and the number of vehicles in the traffic. Personal and collective driving conditions are also effective

in producing pollution. In the USA, hydrocarbon (HC), carbon monoxide (CO) and nitrogen oxide (NO) pollution from transportation is 34.3%, 62.6%, and 34.3% of the total pollution, respectively (De Nevers, 1995).

Theoretical emissions obtained from complete combustion of hydrocarbon fuels (fossil fuels) are CO₂ and H₂O. However, in practice it is impossible to have 100% complete combustion in gasoline and diesel engines. Therefore, some compounds are produced due to incomplete combustion. Exhaust gasses from motor vehicles contain "incomplete combustion products (ICP)" like aldehydes, ketones, carboxylic acids and CO, NO_x, SO₂, lead compounds, particulate matters (Perkins, 1974; Wark and Warner, 1990; Muezzinoğlu, 2004; İcingür, 1991).

The pollutants have a direct and toxic effect on human health. Therefore, emissions from motor vehicles are more important than the other sources. Besides some of these emissions are carcinogenic. Nonmethane hydrocarbons emitted to the atmosphere have an important role on the "Photochemical Smog" formation (Stern et al., 1994).

BTEX is the term used to describe a group of chemicals (hydrocarbons) related to benzene. This includes a variety of compounds: toluene (methylbenzene), ethyl benzene, xylenes and benzene itself. The International Agency for Research on Cancer has designated the components of BTEX as carcinogen (e.g. benzene), possible carcinogen (e.g. ethylbenzene) and not classifiable as to their carcinogenicity to humans (toluene, xylene). However, exposure to BTEX at normal background levels is unlikely to have any adverse effect on human health (SEPA, 2004).

1.2. Objective of the Study

There has been a rapid increase in the number of passenger cars, buses, and shared taxis (dolmuş) during recent years in Turkey. Because of this rapid increase in number of cars during the last 15 years, air quality in big cities has been affected adversely from this situation. In Turkey, no data exists showing the composition of exhaust gasses for different categories of cars. These emissions mainly depend on the: type of fuel used, velocity of the car, driving habits, etc.

Also, there are no emission factors available for cars. Corinair or U.S. EPA derived emission factors are used for estimating traffic-caused emissions. However, these factors do not fit correctly to Turkish car fleet. Therefore, the objectives of this study are to measure the non-methane volatile organic carbon (NMVOC's) emissions from passenger cars in Turkey having gasoline engines, to determine emission factors of these vehicles for BTEX compounds and comparison of emission factors obtained in this study with the emission factors of the other countries.

3. MATERIALS AND METHOD

This study was conducted in two parts. These parts were:

- 1- a) to determine the distribution of vehicle categories in Ankara and Turkey, and choose the category widely used in Turkey.
- 2- To take gas samples from the exhaust pipes of the widely used car types at different road conditions and analyze the gas composition for BTEX components. Calculate the emission factors for the BTEX compounds.

3.1. Experimental

In order to determine the composition of exhaust gasses, samples were collected from the tail pipe and benzene, toluene, xylene, ethylbenzene, m-xylene, o-xylene and 1,2,4 trimethylbenzene (pseudocumene) compounds analyzed by using GC. The type of fuel used was not variable in the experiments. Only the super-unleaded gasoline sold at the gas stations was used. This gasoline is supplied from Middle Anatolia Refinery located close to Ankara. The driving habits were not also included as a variable, because driving at certain constant velocities were considered. Therefore, the only variable was taken as the velocity of the car. The velocities tested were 30 km/hr, 60 km/hr and 90 km/hr corresponding to the urban, rural and highway driving conditions.

The experimental set up consisted of a car, a gas sampling probe to take samples from the tail pipe, a drier, an active carbon tube (Orbo-32) and a suction pump with an adjustable suction rate. A schematic diagram of the car with the experimental set up is shown in Figure 1 and the flow diagram of the exhaust gas collection system is shown in Figure 2. The gas samples from the tail pipe were collected while the car was going at certain speed limits.

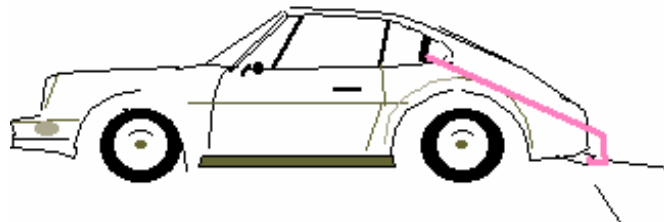


Figure 1. The experimental set up for exhaust gas collection

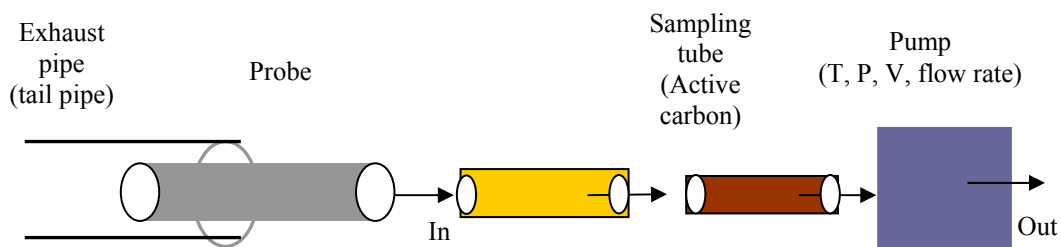


Figure 2. The flow diagram of the exhaust gas collection system

The sampling probe was placed in the tail pipe of the car. About 100 mm of the probe should be in the tail pipe. The probe is made of stainless steel having an inside

diameter of 10 mm and a length of 500 mm. The end of the stainless steel probe is flexible to make a bend about 90°. The outlet of the probe is connected with a PTFE tube to a drier. Drier is made of a glass tube with a diameter of 10 mm. The inside of drier is filled with silica gel beads to adsorb moisture. Both ends of the drier tube are plugged with quartz wool. An Orbo-32 tube containing active carbon particles is placed after the drier tube in the sampling system. The outlet of the Orbo-32 tube is connected to a suction pump (Desega-GS 312) with a PTFE tube. The pump is kept in the car during driving and sampling. All other parts of the experimental set up are outside the car attached to the back bumper. The pump works with 12 V and the power supply of the pump is connected to the battery of the car. Active sampling method was used for gas sampling. VOCs adsorbed on the active carbon particles in the Orbo-32 tube were extracted by liquid CS₂ in the laboratory after the sampling is over. The extraction procedure was followed according to EN/ISO DIS 13528-3 and the components of the extract were analyzed by using a GC having a FID.

The statistical distribution of vehicle categories in Turkey according to the results of the first part of this study show that the cars named under “Murat (Tofas) and Fiat” constitute 34.7% and Renault cars constitute 29.7% of total cars. Therefore, Tofas/Fiat cars were taken for our further studies, because these are widely used cars in Turkey.

In this study passenger cars were divided into mainly two basic categories: 1) Cars without catalytic converter, 2) Cars with catalytic converter. Cars without a catalytic converter are about 10-year old cars since the manufacturing of cars with catalytic converter has started in year 1995. Cars with a catalytic converter are about 4-5-year old cars. The cars chosen for this study without catalytic converter are: Murat-Kartal SLX (1993), Fiat-Tipo (1993), Murat-Dogan SLX (1996); and with catalytic converter are: Murat Kartal (2001), Fiat Marea (2005), Fiat Palio (2000).

Each group of vehicles was examined for three different conditions: cold start (at idle condition), driving condition, and hot start (at idle condition).

The method used during calculation of the "emission factors" at driving conditions is to select one single average speed, representative of each of the road types "urban", "rural" and "highway" and calculate the emission factors at these speeds.

4. RESULTS AND DISCUSSIONS

The samples collected on active charcoal particles in Orbo-32 tubes were analyzed for BTEX compounds. Overall, average exhaust gas VOC concentrations of cars investigated without catalytic converter for three speeds are shown in Figure 3 and with catalytic converter in Figure 4, respectively. As can be seen from these figures, the average exhaust gas VOC concentrations for cars without cat/converter were about 4-6 times more than the average exhaust gas VOC concentrations of cars with cat/converter. The highest concentrations belonged to toluene and m-xylene. When toluene and m-xylene concentrations were 230 and 300 mg/m³ on the average,

respectively, for cars without cat/converter, the corresponding concentrations for the same compounds were 50 and 55 mg/m³, respectively, for cars with cat/converter.

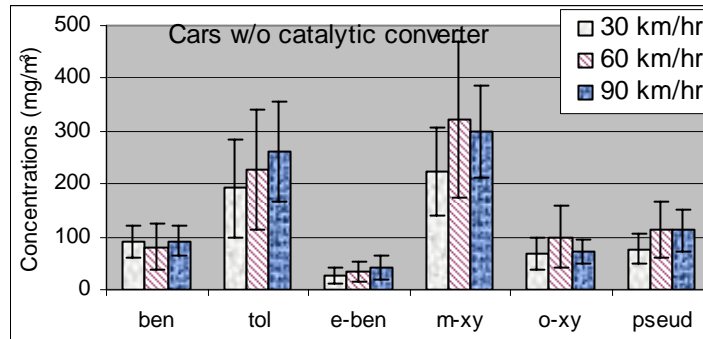


Figure 3. Average exhaust gas VOC concentrations of cars without cat/converter

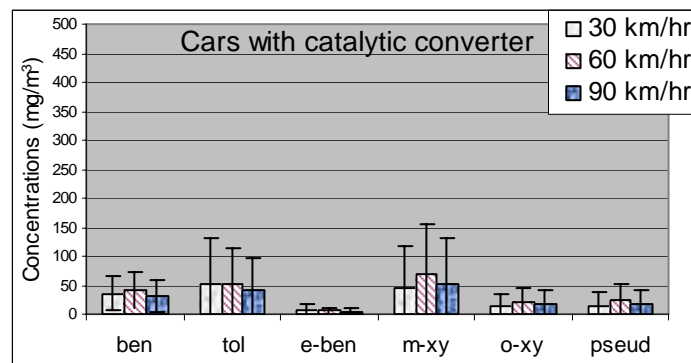


Figure 4. Average exhaust gas VOC concentrations of cars with cat/converter

Average exhaust gas VOC concentrations at idle condition for cars without catalytic converter and with catalytic converter are shown in Figure 5 and 6, respectively. Again the highest concentrations belong to toluene and m-xylene for the idle-cold start and idle-hot start conditions. However, there is not a big difference between the cars with cat/converter and without cat/converter.

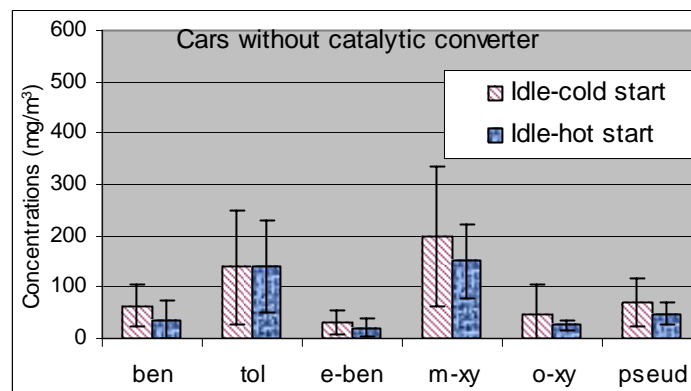


Figure 5. Average exhaust gas VOC concentrations of cars without cat/converter

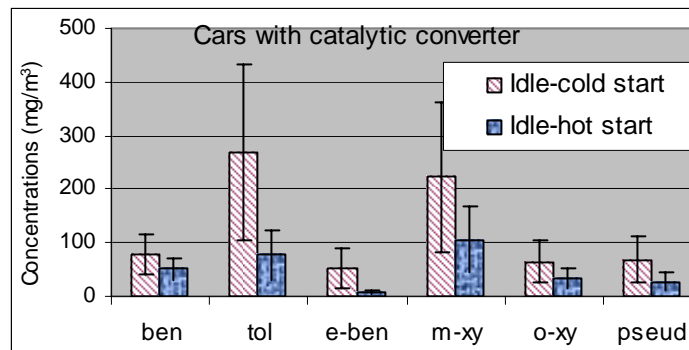


Figure 6. Average exhaust gas VOC concentrations of cars with cat/converter

In general, for cars both with catalytic converter and without catalytic converter the VOC emissions at idle-cold start condition are more than the emissions at idle-hot start condition. In cars without catalytic converter the difference is not much. But for cars with catalytic converter the difference becomes sometimes 2-3 times. For both groups of cars, the largest VOC emission occurs for toluene and m-xylene. The emissions of benzene, e-benzene, o-xylene and pseudocumene are almost at the same order of magnitude.

4.1 Calculation of emission factors

Average emission factors for cars without catalytic converter

The results obtained from the first group of three cars were used to calculate the average emission factors (EF) for cars **without catalytic converter**. These cars had approximately the same cylinder volume and the models were compatible with each other. Emission factors for each road condition as urban, rural and highway driving represented by 30 km/hr, 60 km/hr and 90 km/hr, respectively, were calculated by taking the average of the emission factors of 3 cars at the specified speed. The results are given in Table 1.

Table1. Emission factors of cars without catalytic converter

Parameter	30km/hr	60 km/hr	90 km/hr	Idle cold start	Idle hot start
	EF g/km			EF g/hr	
Benzene	0.08±0.04	0.03±0.01	0.02±0.01	1.34±0.95	0.69±0.66
Toluene	0.20±0.09	0.09±0.05	0.07±0.03	2.88±2.10	2.86±0.50
e-benzene	0.03±0.01	0.01±0.01	0.01±0.01	0.65±0.51	0.40±0.29
m-xylene	0.22±0.09	0.13±0.07	0.09±0.04	4.34±3.44	3.09±0.92
o-xylene	0.08±0.03	0.04±0.03	0.02±0.00	1.10±1.50	0.53±0.85
Pseud.	0.07±0.03	0.04±0.02	0.03±0.02	1.49±1.06	1.01±0.52

EF: Emission factor Pseud: Pseudocumene

It can be easily seen from Table 1 that emission factors generally decrease as the speed increases from 30 km/hr to 90 km/hr. This is attributed to the better

combustion efficiency at higher speeds. For each driving condition emission factors for toluene and m-xylene have been found to be higher than the other VOC compounds investigated, which was an expected result due to the composition of the fuel used.

At idle-hot start condition emission factors were found to be less than idle-cold start condition. This result was also expected because at cold start condition, car's engine has not reached yet its normal working temperature.

Average emission factors for cars with catalytic converter

The experimental results obtained from the second group of three cars were used to calculate the average emission factors (EF) for cars **with catalytic converter**. These cars had approximately the same cylinder volume and the models were compatible with each other. The results for the speeds of 30, 60, 90 km/hr, and for cold and hot start conditions are given in Table 2.

Table 2. Emission factors of cars with catalytic converter

Parameter	30km/hr	60 km/hr	90 km/hr	Idle cold start	Idle hot start
	EF g/km			EF g/km	
Benzene	0.032±0.018	0.012±0.010	0.009±0.007	1.28±0.61	1.01±0.08
Toluene	0.055±0.051	0.015±0.021	0.010±0.012	4.29±2.66	1.25±0.74
e-benzene	0.008±0.007	0.002±0.002	0.001±0.001	0.86±0.61	0.25±0.15
m-xylene	0.049±0.047	0.020±0.030	0.013±0.017	3.57±2.24	1.50±1.32
o-xylene	0.014±0.014	0.006±0.009	0.004±0.005	1.04±0.62	0.67±0.14
Pseud.	0.016±0.015	0.006±0.009	0.004±0.005	1.09±0.68	0.46±0.25

EF: Emission factor Pseud: Pseudocumene

From Table 2, it can be easily seen that emission factors at lower speeds, e.g. 30 km/hr, was higher than the emission factors at higher speeds, e.g. 90 km/hr. For each driving condition emission factors for toluene and m-xylene have been found to be higher than the other VOCs, as expected. At idle-hot start condition emission factors also were found to be less than the idle-cold start condition. This result was also expected as in the previous case.

Results of the statistical analysis which are given in Table 1 and Table 2 show that emission factors found in this study for both cars without catalytic converter and with catalytic converter have large standard deviations from its average values. In order to decrease the standard deviations more data is needed. Therefore, emission factors obtained in this study can only be used to have an idea about the exhaust gas emission factors in Turkey. However, these emission factors in exhaust gas emission calculations should be used carefully.

It can be seen from Tables 1 and 2 that emission factors of cars without catalytic converter were up to 10 times higher than the emission factors of cars with catalytic converter. It can also be seen that generally emission factors from 30 km/hr to 90

km/hr have decreased, and emission factors for cars having catalytic converter are less than that of cars without catalytic converter. Also emission factors for toluene and m-xylene for all cars are higher than the other VOC components measured.

4.2. Total Hydrocarbon (HC) Analysis

Total HCs were analyzed in the exhaust gases of Marea and Palio at idle-hot and idle-cold start conditions. Results of the measurements are given in Table 3.

Table 3. Total HC concentrations of idle-hot start and cold start emissions

Car Type	Total HC Concentration (ppmv)	
	Idle (cold start)	Idle (hot start)
Car 5 (Marea)	463.99±58.35	426.02±73.18
Car 6 (Palio)	1598.89±72.65	1406.72±205.5

It can be seen from Table 3 that total HC concentrations at idle-cold start condition was more than that of idle-hot start condition. HC emissions of Car 5 (Marea) were less than that of Car 6 (Palio). This was an expected result because Marea has a better technology than Palio, and Marea was used less than the other one.

4.3. Comparison of the Results with Literature

Total HC concentrations of Marea and Palio cars in this study were higher than the results obtained in Üner's study (2000). Results of HC concentrations of this study and that of (Üner et al., 2000) which are given in Table 4 were close to each other, although (Üner et al., 2000) studied one month's data and we have studied one year's data. The results of the study are a little higher than Üner's results.

Table 4. Average HC concentrations (Üner et al. (2000) and this study)

Type of cars	Average HC (ppmv) (this study)	Average HC (ppmv) (ÜNER et al., 2000)
TOFAS	298.75±192.64	235.44±80.51
RENAULT	237.66±142.52	221.46±61.51

Emission factors of UK-NAEI (National Atmospheric Emission Inventory) - 2000 and that of this study for light gasoline vehicles are compared in Table 5.

Table 5. Emission factors for benzene (UK-NAEI, 2000 and this study) (<http://www.naei.org.uk/emissions/>, 2004)

Driving condition	UK-NAEI (2000)	Cars w/o catalytic converter (this study)	Cars with catalytic converter (this study)
Benzene emission factors (g/km)			
Urban driving	0.01	0.08	0.032
Rural driving	0.00696	0.03	0.012
Highway driving	0.00727	0.02	0.009

As we compare the emission factors of cars in this study with the literature values, emission factors of benzene are 0.01 g/km for urban driving, 0.00696 g/km for rural driving- 0.00727 g/km for highway driving for UK. These values in this study were calculated as 0.08, 0.03, 0.02 for cars without catalytic converter and 0.032, 0.012 and 0.009 g/km for cars with catalytic converter, respectively. Emission factors of this study for cars without catalytic converter are about 3 to 8 times, and emission factors of cars without catalytic converter are about 1 to 3 times higher than the UK-NAEI emission factors. These differences are thought to be mainly due to the high aromatic content of gasoline used and the vehicle technology available in Turkey.

As we compare the emission factors at idle condition, it can be easily seen that emission factors found in this study were more than the USA road data of 2000 emission factors. According to this data, VOC emission factor for light-duty gasoline-fueled vehicles (gasoline fueled passenger cars) is 0.352 g/hr in winter and 0.269 g/hr in summer conditions. The values in this study are 1.34 g/hr for idle-cold start and 0.69 g/hr for idle-hot start condition for benzene which are three times higher than emission factors of the USA road data. It can easily be said that this result mainly comes from the high aromatic content of gasoline used in Turkey. These values are 1.27 g/hr and 1.01 g/hr for cars with catalytic converter.

5. CONCLUSIONS AND RECOMMENDATIONS

The largest VOC emission occurs for toluene and m-xylene for both car groups (cars with catalytic converter and without catalytic converter). The emissions of benzene, e-benzene, o-xylene, pseudocumene are almost at the same order of magnitude. BTEX components of both types of gasoline are not different from each other. However, concentration and emission factor values of cars without catalytic converter were higher than the concentration and emission factor values for the cars with catalytic converter.

Emission factors for all types of cars are higher for m-xylene and toluene than the other compounds, and decrease as the driving speed increases from 30 km/hr to 90 km/hr.

Emission factors calculated in this study were higher than the emission factors found in the literature. These differences are mainly due to the high aromatic content of gasoline used in Turkey. Today, Ministry of Environment and Forestry is also trying

to reduce emissions coming from these sources by reducing aromatic content in gasoline and encouraging the usage of new technology for car manufacturing.

It is better to analyze HC's in g/km to be comparable with the EU emission factors. Results of this study showed that exhaust emissions of cars are very high at 30 km/hr velocity which is mainly used in crowded streets and busy intersections. Therefore, exhaust emissions are more during stop-and-go traffic. In order to decrease these emissions synchronization of traffic lights, underpass and overpasses are recommended in crowded streets and busy intersections to decrease the stop-and-go traffic, and the exhaust emissions.

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AIR POLLUTION SHORT CIRCUIT EFFECTS OF ROAD TRAFFIC TUNNEL PORTALS

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ABSTRACT

In the design of road traffic tunnels, the air quality inside of traffic tunnels is an important factor, especially for long road traffic tunnels. The air quality inside the traffic tunnels usually depends on the emission of the cars passing through the traffic tunnel and the rate of tunnel ventilation. In specific situations the concentration of air pollution in traffic tunnels depends also strongly on aerodynamic short circuit (unwanted recirculation) effects that occur around the tunnel portals. At certain meteorological conditions (wind speeds and wind directions), due to the influence of wind vortexes, induced by the tunnel portal geometry, a significant part of the exhaust gasses, blown out of the tunnel portal of the outgoing traffic, is transported to the tunnel portal used by ingoing traffic. Even upstream effects, i.c. air pollution effects upwind, can be seen. Without measures the concentrations of air pollution in the traffic tunnel can reach very high levels.

In this study these effects are investigated for a new to be built road traffic tunnel. This tunnel with a total length of approximately 800 m, consists of 2 major tunnel ducts, a four-lane duct and a two-lane duct, separated from each other by a closed wall. The four-lane duct is for one-way traffic in northern direction. The two-lane duct will be used during the morning rush hour for traffic in southern direction and in the evening rush for traffic in northern direction. Therefore, during the morning rush there will be traffic in opposite directions in the two ducts of the new tunnel. In these situations short circuit effects of air pollution coming from one tunnel duct to the other are known to take place, depending on the meteorological conditions and the design of the tunnel portal. The amount of short cut defines the amount of ventilation needed to control air quality inside the tunnel and should therefore be reduced as far as possible.

In order to determine the amount of air pollution short circuit, a scale-model (1:150) of the tunnel was built. The short circuit effects near the tunnel portals were thoroughly researched, using tracer gas measurements executed in the Peutz atmospheric boundary layer windtunnel in Mook in the Netherlands. For different meteorological conditions, the influence of different tunnel portal designs on the rate of short circuit was determined. Investigated were the influences of tunnel portal canopy, smoke fences and extended (individual) tunnel portals. In worst case situations the rate of air pollution short circuit can be over 50%, which means that the concentration of the air pollution transported to the ingoing tunnel portal, can be higher than 50% of the concentrations of the emitted air pollution of the outgoing tunnel portal. Based on the windtunnelstudy, a specific tunnel design can be made in

order to minimise the short circuit effects. With a good tunnel portal design the rate of air pollution short circuit effects can be limited to 10%. The results of the concentration measurements performed in the windtunnel were illustrated using smoke dispersion tests in the windtunnel, which helps to understand the measured phenomena.

Key Words: Air Pollution, Road Traffic Tunnel

1. INTRODUCTION

Peutz bv performed wind tunnel research into the air pollution (aerodynamic) short circuit occurring at the tunnel portals of a planned road traffic tunnel under a sea canal. The existing road traffic tunnel is the tunnel through which the motorway passes under the sea canal. The existing 2 x 2-lane tunnel is being stretched to the limit by the volume of traffic during the (morning and evening) rush hours. Therefore a new road traffic tunnel is planned to the east of the present road traffic tunnel.

The planned road traffic tunnel has a total length of approximately 800 m and will consist of a tunnel with a total of 6 traffic lanes, i.e. a western tube with 2 lanes and an eastern tube with 4 lanes. The new 2-lane road traffic tunnel will be used during the morning rush hour for traffic travelling south. In this situation it is possible, owing to traffic moving in opposite directions in tubes located alongside one another, that aerodynamic short circuit may occur. Figure 1 (mouth of the tunnel south) and figure 2 (mouth of the tunnel north) show the situation of both tunnel portals of the new road traffic tunnel.

This research concentrates on the air pollution short circuit occurring with the new road traffic tunnel. The research provides insight into the effects of a range of structural variants for the tunnel portals on the air pollution short circuit at the tunnel portals. As an indicator for the aerodynamic short circuit occurring, use was made of the short circuit percentages determined by means of wind-tunnel research.

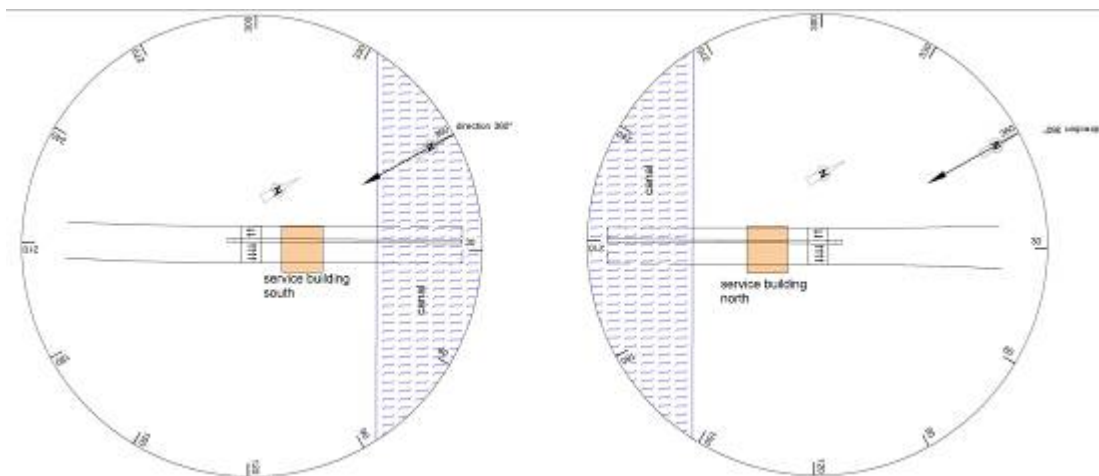


Figure 1: Road Traffic Tunnel.
Situation South, 2 lanes out, 4 lanes in.

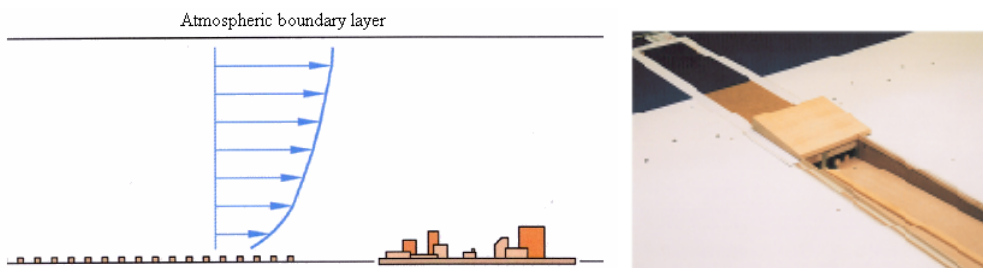
Figure 2: Road Traffic Tunnel.
Situation North, 2 lanes in, 4 lanes out.

2. STARTING POINTS

The planned road traffic tunnel will consist of a tunnel with a total of 6 traffic lanes, i.e. a western tube with 2 lanes and an eastern tube with 4 lanes, see figure 1 and 2. Following realisation of the road traffic tunnel, the traffic travelling south will pass through the present road traffic tunnel (2 x 2-lane) and traffic travelling north through the 4-lane tube of the new road traffic tunnel. The 2-lane tube of the new road traffic tunnel will be used during the morning rush hour for traffic travelling south and during the evening rush hour for traffic travelling north. This "variable direction of travel" means that a total of (existing tunnel and the additional new road traffic tunnel) a minimum 4 and a maximum of 6 traffic lanes will be available, both in the southerly and northerly direction, allowing traffic flow to be improved.

If the 2-lane tube of the new road traffic tunnel is used for traffic travelling south (morning rush hour), it is possible that traffic moving in opposite directions in adjacent tubes could lead to aerodynamic short circuit occurring. This means that some of the traffic emissions to the air, caused by the traffic emerging from the tunnel, can be directly taken into the other tube by the traffic driving into that tunnel (due to both the wind and ventilation and the movement of the traffic). These effects depend on the meteorological conditions (wind speed and wind direction) and the tunnel portal geometry. In order to determine the effect of air pollution short circuit a scale model (1:150) of the road traffic tunnel was built to test in the atmospheric windtunnel. Figure 3 shows the atmospheric boundary layer and a model of the tunnel portal. The average (constant) air velocity in the tube of the new road traffic tunnel should be 5 m/s (ventilation design criteria). The aerodynamic effect nearby the tunnel portal depends strongly on the relation between the air velocity in the road traffic tunnel and the (free field) wind speed at 10 metres high, so the research is made for different speed ratios.

Figure 3: Atmospheric boundary layer and scale model of the tunnel portal.



3. WINDTUNNEL STUDY

Figure 4 shows the atmospheric boundary layer windtunnel, the model and surrounding equipment.

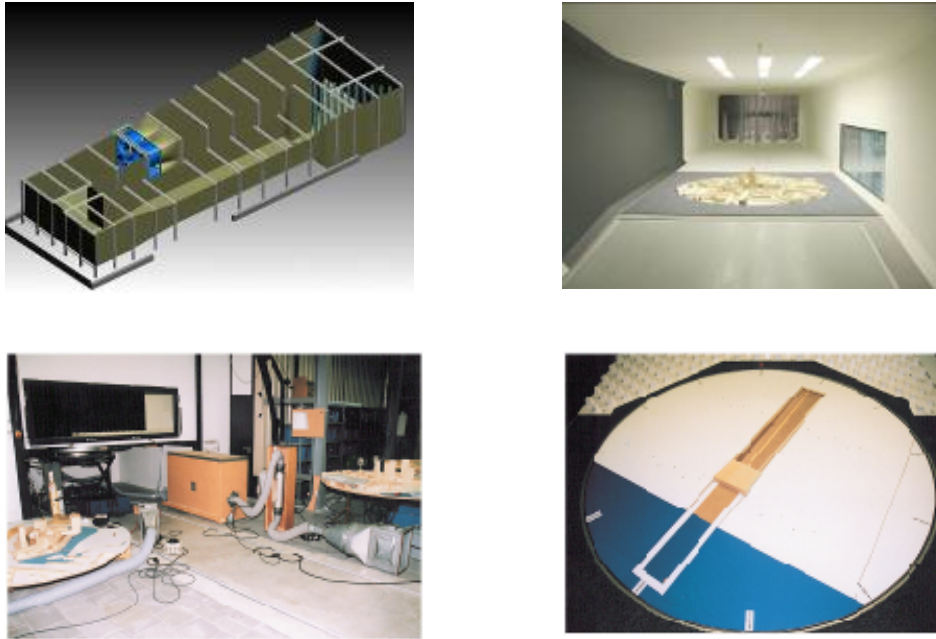


Figure 4: Peutz atmospheric boundary layer windtunnel, model and surrounding equipment.

For the purposes of this research into the road traffic tunnel, the situation with 4-lane traffic in a northerly direction and 2-lane traffic in a southerly direction is relevant (morning rush hour). For the northern tunnel portal, the 4-lane exiting and 2-lane entering situation was therefore taken. For the southern tunnel portal, the situation considered was with 2 lanes of exiting traffic and 4 lanes of traffic entering the tunnel.

In the possible situation with 6 lanes of traffic travelling in a northerly direction (evening rush hour), there would be no traffic travelling in the opposite direction and therefore no aerodynamic short circuit occurring.

The concentration measurements for the flow research were carried out using tracer gas (isobutylene) in combination with PID monitors. The measured concentrations were being corrected for background concentrations.

Variant research

The following structural variants were considered in this research (for the north portal of each variant, a photo of the modified scale model is included, see Figures 5 through 10):

- *Variant A: Present design (Figure 5)*
Smoke barrier with canopy (canopy 50% open).

- Length smoke barrier approx. 20 metres;
- *Variant B: Present design without canopy (Figure 6)*
Without smoke barrier and canopy, central reservation with wall approx. 1 metre high;
 - *Variant C1: Present design with raised smoke barrier (Figure 7)*
As A, in addition smoke barrier raised to ground level;
 - *Variant C2: Present design with lengthened and raised smoke barrier (Figure 8)*
As C1, raised smoke barrier with approx. 20 metres of extra length (total approx. 40 metres);
 - *Variant D1: Present design with extended exit (Figure 9)*
As A, in addition tube exit extended by approx. 20 metres;
 - *Variant D2: Present design with extended exit extension (Figure 10)*
As D1, tube exit extended by a further approx. 20 metres (total approx. 40 metres);

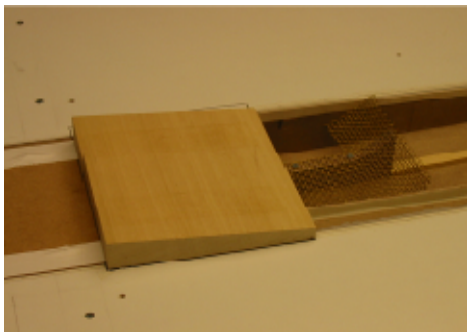


Figure 5 (variant A)

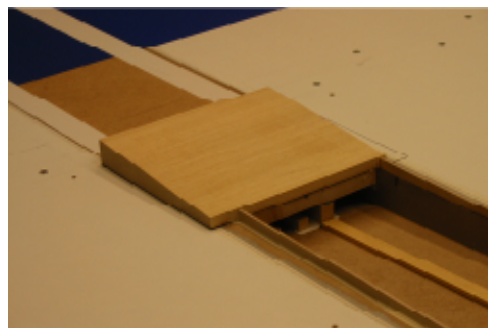


Figure 6 (variant B)

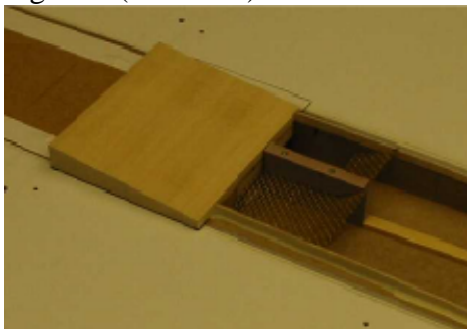


Figure 7 (variant C1)

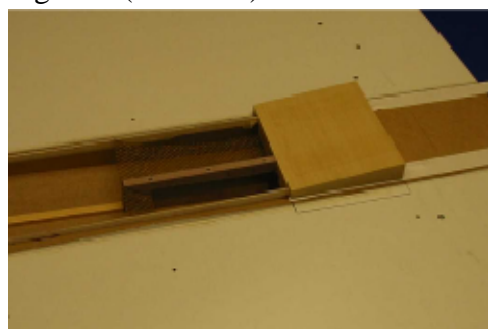


Figure 8 (variant C2)

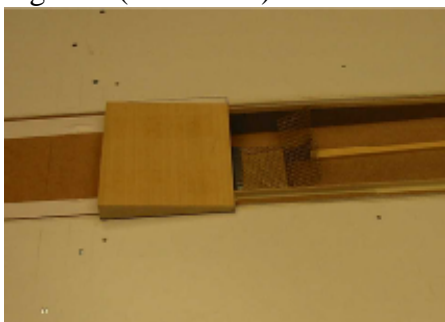


Figure 9 (variant D1)

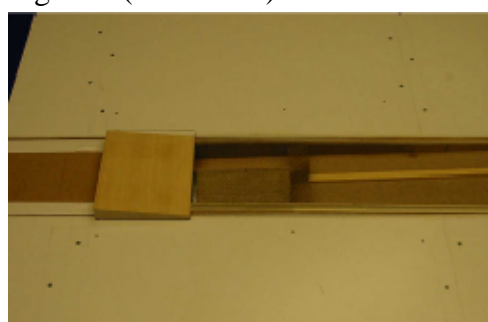


Figure 10 (variant D2)

The extent of air pollution short circuit is determined in part by the relationship between the ventilation speed in the tube (V_t , constant 5 m/s) and the wind speed in

the environment at 10 metres high (V_w , variable). For the purposes of this research, the following 3 speed ratios were applied:

$V_w / V_t = 2$ wind speed 10 m/s, ventilation speed 5 m/s

$V_w / V_t = 1$ wind speed 5 m/s, ventilation speed 5 m/s

$V_w / V_t \sim 0$ wind speed ~ 0 m/s, ventilation speed 5 m/s

It should be however noted that, owing to measuring circumstances, the lowest speed ratio was approximated by applying $V_w = 0.6$ m/s at a V_t of 5 m/s (in fact $V_w / V_t = 0.12$).

This wind-tunnel research therefore involved 36 variants, i.e. 6 structural variants for 3 different speed ratios for both tunnel portals. To verify the measurements, a set of control measurements were performed (reproducibility).

Measurement results air pollution short circuit

The concentration measurements in the wind-tunnel resulted for each structural variant, each speed ratio (V_w / V_t) and each wind direction in an emission concentration at the exit (representing concentration of exhaust gasses resulting from the traffic exiting the tunnel tube) and an immission concentration at the entrance of the other tube. The immission concentrations were determined downstream in the tube. In figure 11 an example of a measured situation is given. The measurements revealed that the aerodynamic short circuit occurring at the speed ratio $V_w / V_t \sim 0$ is virtually unaffected by the wind direction, owing to the relatively low wind speed.

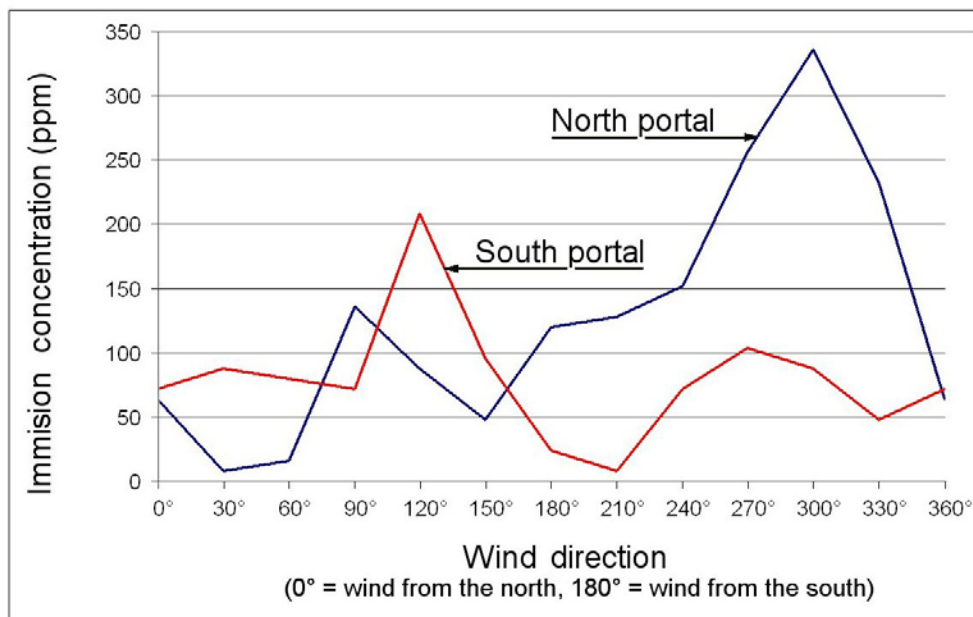


Figure 11: Example of measured immission concentration (ppm) downstream in road traffic tunnel tube as a function of wind direction (°): $V_w / V_t = 2$, variant A, emission concentration from emitting tube approximately 800 ppm.

4. CALCULATIONS

4.1. Calculation of air pollution short circuit

For a given wind speed and wind direction, the emission concentration at the exit and the immission concentration downstream from the entrance can be used to calculate the short circuit percentage. The formula below is used for this:

$$R = \frac{C_{immission}}{C_{emission}} \cdot 100\% \quad [1]$$

Whereby:

- R = short circuit percentage [%]
- C_{emission} = emission concentration exit [ppm];
- C_{immission} = immission concentration entrance (downstream, corrected for background concentration) [ppm].

4.2. Results of the calculations for short circuit

The short circuit percentage is determined on the basis of the emission and immission concentrations found for each measurement, using formula [1]. The minimum and maximum air pollution short circuit percentages occurring are given in table 1.

Table 1: minimum and maximum air pollution short circuit percentages for several design variants; tunnel portal north and south.

Tunnel portal north						
	V _w = 10 m/s, V _t = 5 m/s		V _w = 5 m/s, V _t = 5 m/s		V _w = 0,6 m/s, V _t = 5 m/s	
variant*	minimum	maximum	minimum	maximum	minimum	maximum
A	ca. 1%	ca. 42%	ca. 5%	ca. 17%	ca. 1%	ca. 6%
B	ca. 8%	ca. 53%	ca. 8%	ca. 35%	ca. 10%	ca. 12%
C1	ca. 1%	ca. 34%	ca. 2%	ca. 19%	ca. 1%	ca. 2%
C2	ca. 0%	ca. 20%	ca. 1%	ca. 17%	ca. 1%	ca. 2%
D1	ca. 1%	ca. 32%	ca. 1%	ca. 20%	ca. 0%	ca. 1%
D2	ca. 0%	ca. 18%	ca. 1%	ca. 13%	ca. 1%	ca. 1%

Tunnel portal south						
	V _w = 10 m/s, V _t = 5 m/s		V _w = 5 m/s, V _t = 5 m/s		V _w = 0,6 m/s, V _t = 5 m/s	
variant*	minimum	maximum	minimum	maximum	minimum	maximum
A	ca. 1%	ca. 26%	ca. 4%	ca. 15%	ca. 2%	ca. 3%
B	ca. 6%	ca. 39%	ca. 7%	ca. 27%	ca. 8%	ca. 10%
C1	ca. 2%	ca. 21%	ca. 2%	ca. 15%	ca. 1%	ca. 2%
C2	ca. 1%	ca. 14%	ca. 2%	ca. 15%	ca. 1%	ca. 4%
D1	ca. 2%	ca. 22%	ca. 2%	ca. 14%	ca. 1%	ca. 2%
D2	ca. 2%	ca. 12%	ca. 1%	ca. 10%	ca. 1%	ca. 1%

4.3. Reproducibility

The accuracy of the concentration measurements is approx. 5 ppm, which leads to a level of inaccuracy in the short circuit percentages of 1 to 2%.

A comparison of the short circuit percentages from control measurements with the short circuit percentages from the standard measurements shows that these can be reproduced with sufficient accuracy (+/- 2%).

5. CONSIDERATIONS

From the measurements and calculations made for the planned road traffic tunnel, it appears that the maximum air pollution (aerodynamic) short circuit occurs at a speed ratio of $V_w / V_t = 2$. The short circuit occurring at the other speeds tested ($V_w / V_t = 1$ and $V_w / V_t \sim 0$) is generally (considerably) lower.

At the north portal, the maximum air pollution short circuit occurs with a north-westerly wind (300°). At the south portal, the maximum aerodynamic short circuit occurs with a south-easterly wind (120°). This corresponds to expectations in this respect; the occurrence of "slipstream effects" at the mouth of the tunnel in the tunnel trough in these situations means that a significant portion of the emissions from the 4-lane tube is sucked back into the 2-lane tube.

Table 1 shows that at the north portal, with variant A (present design), the aerodynamic short circuit would reach a maximum of approx. 42%. At the south portal, the short circuit in this variant would be a maximum of approx. 26%.

In the case of variant B (present design without canopy), the air pollution short circuit would reach a maximum of approx. 53% at the north portal and approx. 39% at the south portal.

In the case of C1 (present design with raised smoke barrier), the air pollution short circuit would reach a maximum of approx. 34% at the north portal and approx. 21% at the south portal.

In the case of C2 (present design with extended and raised smoke barrier), the air pollution short circuit would reach a maximum of approx. 20% at the north portal and approx. 14% at the south portal.

In the case of D1 (present design with extended exit), the air pollution short circuit would reach a maximum of approx. 32% at the north portal and approx. 22% at the south portal.

In the case of D2 (present design with extended exit extension), the air pollution short circuit would reach a maximum of approx. 18% at the north portal and approx. 12% at the south portal.

The above considerations per variant show that the maximum short circuit occurring at the south portal is in all cases less than that at the north portal, which is a consequence of the situations considered above. For the northern tunnel portal, the 4-

lane exit and 2-lane entrance situation was considered, while for the southern tunnel portal the 2-lane exit and 4-lane entrance situation was considered. The determining slipstream in the former situation is present over a larger polluted area of emission than in the latter, whereby in the former situation a larger portion of the emissions will be sucked into the other tube by means of (aerodynamic) short circuit. This research has revealed that the most air pollution short circuit occurs in variant B (present design without canopy).

Virtually the same amount of short circuit is also present in variants C1 (present design with raised smoke barrier) and variant D1 (present design with extended exit), but less than with variant A (present design). Raising the smoke barrier to ground level would reduce the maximum air pollution short circuit occurring in relation to variant A (present design) by approx. 5 - 8%; extending the exit by approx. 20 metres reduces the maximum air pollution short circuit occurring in relation to variant A (present design) by approx. 4 - 10%, all depending on the tunnel portal under consideration (north or south).

In the case of variant C2 (present design with extended and raised smoke barrier) and variant D2 (present design with extended exit extension), the amount of short circuit is virtually the same, but less than in the case of variants C1 and D1. By extending the raised smoke barrier by approx. 20 metres, the maximum aerodynamic short circuit occurring in relation to variant C1 is reduced by a further approx. 7 - 14%. In relation to variant A (present design), the maximum short circuit occurring is reduced by approx. 12 - 22%. By lengthening the exit by another approx. 20 metres (total approx. 40 metres), the maximum aerodynamic short circuit occurring in relation to variant D1 is reduced by a further approx. 10 - 14%. In relation to variant A (present design), the maximum short circuit occurring is reduced by approx. 14 - 24%.

6. CONCLUSIONS

On the basis of the starting points, the wind-tunnel research and the calculations concerning the aerodynamic short circuit occurring at the tunnel portals of the planned road traffic tunnel, it can be said that:

- the maximum air pollution short circuit occurring for the present design would be approx. 42%;
- if the present design were to be implemented without canopy and smoke barrier, the maximum air pollution short circuit would increase to significant more than 50%;
- raising the smoke barrier (to ground level) would reduce the aerodynamic short circuit in relation to the present design by approx. 5 - 8%, depending on the tunnel portal in question (north or south);
- extending the exit (by approx. 20 metres) would reduce the aerodynamic short circuit in relation to the present design by approx. 4 - 10%, depending on the tunnel portal in question (north or south);
- extending the raised smoke barrier to approx. 40 metres would reduce the aerodynamic short circuit in relation to the present design by approx. 12 - 22%, depending on the tunnel portal in question (north or south);

- extending the exit to approx. 40 metres would reduce the aerodynamic short circuit in relation to the present design by approx. 14 - 24%%, depending on the tunnel portal in question (north or south).

On the basis of the research carried out, insight has been obtained into the various flow effects, such as slipstream effect and thinning so that, taking undesired short circuit effects into account, a well-considered choice of structural variant can be made.

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COMPOSITION AND TEXTURAL PROPERTIES OF SOOT AND STUDY OF THEIR OXIDATIVE ELIMINATION BY CATALYTIC PROCESS

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ABSTRACT

Diesel soot and reference carbon black samples have been characterized by analysis of the organic compounds fraction and determination of textural characteristics (specific area). The potential to eliminate such carbonaceous particles has been investigated using a Cu/ZrO₂ solid catalyst promoted by potassium additives. The reactivity of soot samples depends on its organic compounds content. In general, Cu-K/ZrO₂ catalyst is able to lower significantly the soot oxidation temperature of almost 200°. This high activity is related to the ability of potassium to interact with copper species and to favour the release of active oxygen from the catalyst.

Key Words : soot, surface area, organic fraction, oxidation, Cu-K/ZrO₂ catalysts

1. INTRODUCTION

Soot particles correspond to a major fraction of atmospheric aerosols particularly in urban areas concerned by an important Diesel traffic density. Negative effects of soot are nowadays well-recognized on human health. The particle size appears to be the most influential factor (Schwartz and Meas, 2000) however specific organic compounds in soot could be one of the characteristics explaining that these airborne particles are more bioreactive than others (Adamson et al., 2000). During the last years, the awareness of the impacts of Diesel soot has lead to several works in catalysis research. In fact, among the technologies considered for soot removal, one way consists in the use of catalytic traps, corresponds to filters carrying a suitable catalyst, in view of simultaneous soot filtration and combustion. The catalyst should lower down soot ignition temperature of the non - catalytic combustion to the values characteristic of diesel exhaust: i.e. from almost 600°C to 200-350°C, respectively. The aim of this work is to undertake a study on both the chemical composition of different diesel soot samples and their textural properties (surface areas). These soot samples were then tested in an oxidative elimination reaction in presence of a Cu/ZrO₂ solid catalyst promoted by potassium additives. Different Cu and K

contents were considered for a better understanding of the effect of each active element on the oxidation reaction.

2. EXPERIMENTAL AND METHODS

The organic compounds of different Diesel soot DS (engine operating conditions in both low and high combustion temperatures) and reference carbon black samples (synthetic furnace soot NC330 Degussa and synthetic flame soot Printex U Degussa) were analyzed by chromatographic method coupled to mass spectrometry (GC-MS) for the qualitative/quantitative determination of polycyclic aromatic hydrocarbons and paraffins. Specific areas of soot and carbon black samples were determined by nitrogen adsorption.

The catalysts were prepared by co-impregnation in aqueous medium of given amounts of copper [$\text{Cu}(\text{NO}_3)_2$] and potassium [KHCO_3] onto a ZrO_2 carrier. After drying, all solids were calcined under dry air for 4h at 600°C . The equivalent amounts of CuO or/and K_2O in calcined catalysts were determined by chemical analysis. Catalysts were designated $\text{Cu}_x\text{-K}_y/\text{ZrO}_2$ where $0 < x < 4$ and $0 < y < 4$ correspond respectively to the amount expressed in wt% of CuO and K_2O in catalysts calcined at 600°C .

The catalytic tests were performed using a mixture of soot (6 wt%) and catalyst (94 wt %) corresponding to loose contact conditions (Neeft et al., 1997, Courcot et al., 2004). The latter permit to evaluate the activity of oxide catalysts in experimental conditions closely similar to those of real application, since the soot particles emitted through the exhaust line can only be stopped by the catalyzed filter surface. The catalytic oxidation was studied by simultaneous TG-DSC analysis (NETZSCH STA 409 apparatus). About 50 mg of the CB-catalyst mixtures were loaded in an alumina crucible and heated from 25 to 800°C using a heating rate of 5°.min^{-1} in air flow ($75\text{ml}\cdot\text{min}^{-1}$). As shown in Figure 1a, by processing the experimental data, the onset temperature (T_i) could be derived (i.e. temperature at which the combustion starts). The value of T_i is calculated from the TG curve as the intersection between the baseline and the tangent of the curve traced at the inflection point. Following a similar method, the temperature (T_f) at which the combustion of carbon is complete was obtained.

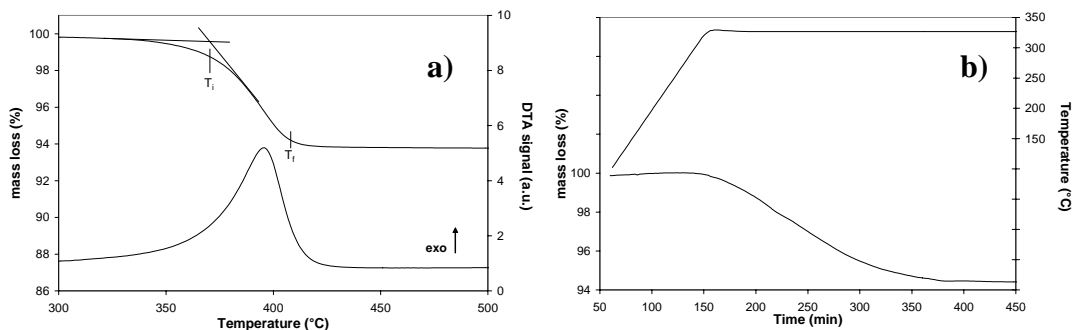


Figure 1 : Examples of thermal analysis curves recorded for the catalytic test of (CB-catalyst) mixtures. a) non-isothermal conditions; b) isothermal conditions.

Additional catalytic tests were performed under isothermal conditions considering the CB-catalyst mixtures in loose contact. Samples were kept under an air flow ($75\text{ml}\cdot\text{min}^{-1}$) and heated to 327°C ($5^\circ\cdot\text{min}^{-1}$). The weight loss rate was used as a measure for the CB oxidation rate (Figure 1b) in agreement with the experimental approach described by van Setten et al. (1999). Considering tangent of the mass curve at different carbon conversion values, oxidation rates were calculated. Once the combustion starts, it is observed that the oxidation rate progressively increases, reaches a maximum and then finally decreases to zero. The analysis of combustion products was performed evaluating the $\text{CO}/(\text{CO}+\text{CO}_2)$ molar ratio from a Varian 3600 chromatograph coupled to the thermobalance.

The reducibility of the catalysts was determined by Temperature Programmed Reduction (H_2 -TPR). The experiments were carried out in a conventional apparatus (AMI 200, Zeton Altamira). The catalysts (~ 75 mg) were previously treated *in-situ* under O_2 diluted in argon flow at 450°C for 1 h. After cooling under argon flow, reduction treatment (3% H_2 in argon) was performed with a total flow of $30\text{ mL}\cdot\text{min}^{-1}$ from 25°C to 700°C with a heating rate of $10^\circ\cdot\text{min}^{-1}$. Additional experiments consisted in H_2 -TPR measurements of (CB-catalysts) loose contact mixtures previously treated under argon ($30\text{ mL}\cdot\text{min}^{-1}$) at 400°C for 1 h. This treatment was considered to simulate the beginning of the CB conversion initiated by the participation of active catalytic species, normally contributing to a redox mechanism. Considering the difference in H_2 consumption for these two kinds of measurements for a given catalyst, quantitative information could be obtained on such oxygen species able to react with carbon during the Ar treatment at 400°C .

3. RESULTS AND DISCUSSION

3.1. Soot and Carbon Black characterization

The analysis of the nine PAHs given in Table 1 indicates that the diesel soot contains the higher content of PAHs. The synthetic flame CB (Printex U) contains higher amounts of PAHs than the synthetic furnace sample (NC 330).

Table 1. Concentration ranges ($\mu\text{g}/\text{g}$) of individual PAH in diesel soot and carbon black samples.

Compounds	Diesel soot DS	CB Printex U	CB NC 330
Phenanthrene	744.8	810.1	6.8
Anthracene	361.2	78.9	-
Fluoranthene	150.6	139.6	5.5
Pyrene	89.8	106.4	20.7
Benzo(a)anthracene	174.2	2.6	-
Chrysene	117.4	4.5	-
Benzo(a+e)pyrene	75.4	18.7	-
Indeno(123cd)pyrene	52.3	2.0	-
Benzo(ghi)perylene	138.1	1.4	-

Table 2. Textural characteristics of Diesel soot and carbon black .

Samples	Specific area (m ² /g)	
	outgassed at 100°C	outgassed at 500°C
Diesel soot DS	43	262
carbon black NC 330	80	82
carbon black Printex U	95	128

Table 2 gathers the specific areas values for samples outgassed at 100°C and 500°C. Except in the case of CB NC 330, a significant increase in specific area occurred after outgassing at 500°C, and particularly in the case of the diesel soot DS sample. More precisely, we observed a specific area increase reaching 73 m²/g after treating the diesel soot at 300°C. Then the main specific area increase is observed 300°C and 400°C, to reach 240 m²/g. Simultaneously, TG-DTA measurements performed under pure N₂ gas reveals a progressive desorption of organic compounds between 200° and 400°C (not shown). The evolution of the soot specific area versus the treatment temperature seems to be correlated to the departure of organics compounds from the soot. In fact, we evidenced the desorption of 8.2 % of volatile compounds between 200°C and 300°C among them alkylated benzene and alkylated naphthalene that are more generally found in the gaseous phase of the exhaust (Flandrin et al. 2002). Others main compounds desorbed were acenaphthylene, acenaphthene and fluorene. Hence the total organic fraction of the diesel soot include volatile compounds and the PAHs given in Table 1. The volatile compounds would be mainly adsorbed on soot surface sites. On the other hand, the nitrogen adsorption isotherm for the soot outgassed at 500°C was typical of a porous material, characterized by a pore width between 20 and 30 nm, obtained from density functional theory model. These data can be explained considering the departure of a part of the heavier organic compounds, probably occupying the inter-particle pore volume of the soot. This result appeared to be in good agreement with findings of Rockne et al. (2002) on the pore structure of soot.

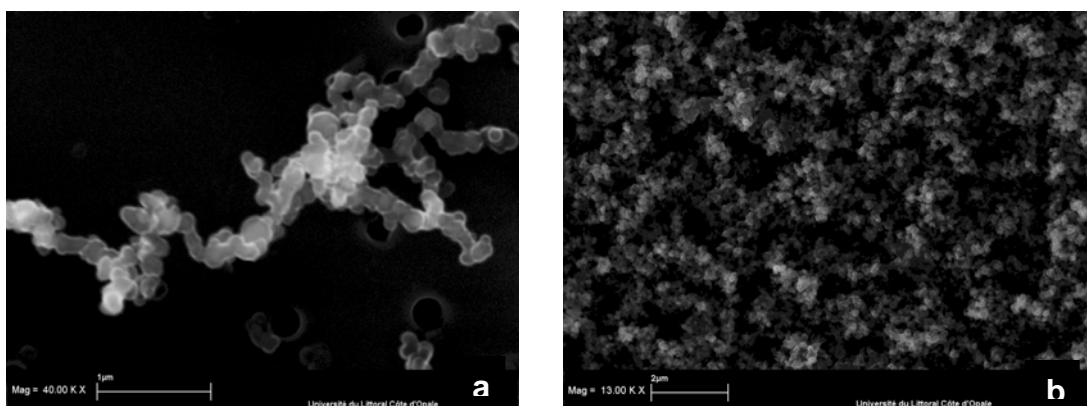


Figure 2: Secondary Electron images of diesel soot particles. a) structured chain with a low agglomeration ; b) highly structured and agglomerated soot.

The SEM analysis of the diesel soot particles (Figure 2) shows a typical chain-like structure of soot (Ambrogio et al. 2002). The latter corresponds to the agglomeration of elemental soot particles, with diameter close to 80 – 120 nm in our case. The soot sample also shows a high level of agglomeration. This high agglomeration can probably be considered to explain the presence of the inter-particle porosity of the soot.

3.2. Soot oxidation by Cu-K/ZrO₂ catalysts

Catalytic tests were first performed using NC 330 CB since this sample contains a low amount of organic compounds and almost only a carbonaceous matrix. In this way, NC 330 can be used a reference to evaluate the ability of a catalyst to oxidize carbon. Figure 2 gives the temperature range in which the oxidation of NC 330 occurred for the test of (NC 330 - catalyst) mixtures. It is important to notice that the oxidation of non-catalyzed NC 330 in air flow occurred regularly between $T_i = 530^\circ\text{C}$ and $T_f = 635^\circ\text{C}$. The results obtained in the presence of pure ZrO₂ indicated the low reactivity of this oxide carrier. The reaction temperature range for Cu_x/ZrO₂ and ZrO₂ exhibits only little difference, revealing a slight improvement in catalytic properties, which depends on the copper content. The potassium oxide species leads to a high improvement of the reactivity of the catalytic systems, also depending on the K content. K_{3,50}/ZrO₂ catalyst permits to lower down soot ignition temperature between $T_i = 385^\circ\text{C}$ and $T_f = 475^\circ\text{C}$. For the catalytic tests performed in the presence of K-Cu/oxide, the conversion of carbon occurred in a low temperature range. In fact, it is noteworthy to obtain the shift toward lower temperatures of nearly 200° between the test performed with Cu_{1,96}-K_{3,45}/ZrO₂ and ZrO₂. The catalyst Cu_{1,96}-K_{3,45}/ZrO₂ ($T_i = 370^\circ\text{C}$ and $T_f = 445^\circ\text{C}$) appeared then as the most reactive system indicating synergistic effects related to the addition of both Cu and K on the ZrO₂ surface.

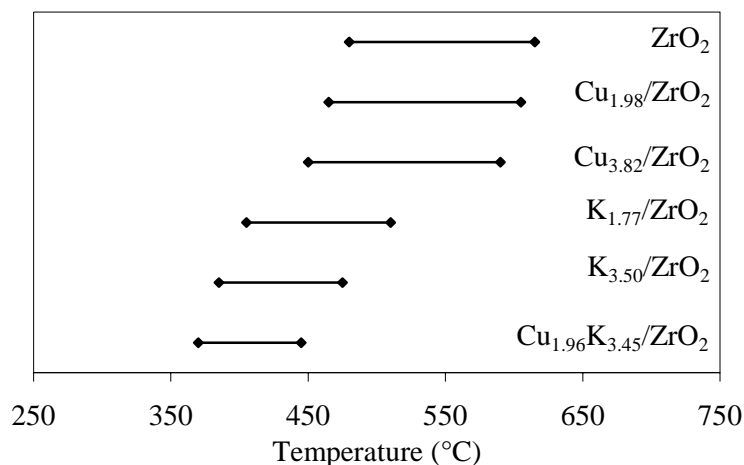


Figure 3 : Temperature ranges for NC 330 oxidation in the presence of Cu_x-K_y/ZrO₂ catalysts

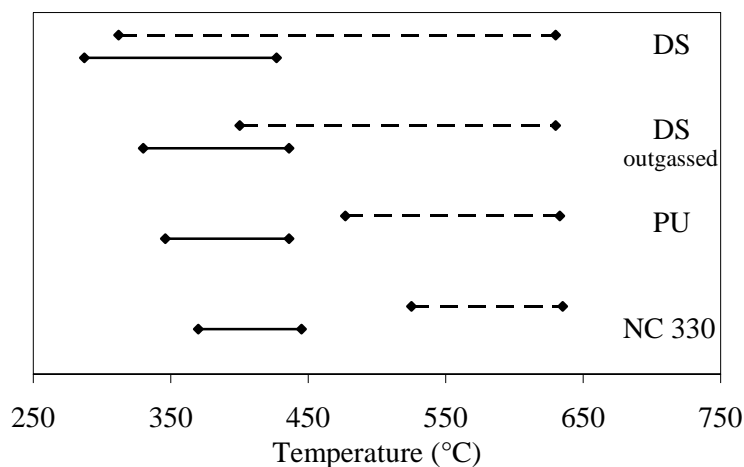


Figure 4 : Temperature ranges of Diesel soot and carbon black samples in the presence of $\text{Cu}_{1.96}\text{K}_{3.45}/\text{ZrO}_2$ catalyst.

The soot and carbon black samples were then tested in the oxidation reaction in the presence of the most active $\text{Cu}_{1.96}\text{-K}_{3.45}/\text{ZrO}_2$ catalyst and the catalytic effect can be appreciated by comparison with the non-catalytic oxidation data. First, the ignition of DS is observed at lower temperature than the DS outgassed due to the occurrence of organic compounds oxidation at a relatively low temperature in comparison with the carbon fraction. In this series, NC 330 appeared as the less reactive carbonaceous compound. It can be outlined that nearly same Tf values ($\sim 630^\circ\text{C}$) were obtained for the different samples, revealing similar properties towards the oxidation of the remaining carbon fraction after the organic compounds or/and surface functions reacted in the lower temperature range. A similar reactivity classification has been obtained when the oxidation is performed in the presence of $\text{Cu}_{1.96}\text{-K}_{3.45}/\text{ZrO}_2$ catalyst. Nevertheless, it appeared that the shift towards lower temperatures is more important on Tf values than Ti values. Considering at first the oxidation of organic compounds at low temperature (close to Ti) and rather the oxidation of carbon at higher temperature, this observation tends to show that such catalysts are more efficient in lowering the oxidation of carbon (almost $180 - 200^\circ$) than organic compounds in soot (between 30 and 150°).

Complementary information on the soot reactivity in the presence of the $\text{Cu}_{1.96}\text{-K}_{3.45}/\text{ZrO}_2$ catalyst were obtained from soot oxidation rates determined under isothermal conditions (Figure 5). The higher oxidation rates values were obtained after soot conversion of 20 or 30% for DS and Printex U, which correspond to samples containing the higher fraction of organic compounds. For these samples, oxidation rates values remain higher than DS outgassed and NC 330 till a conversion of 60% is reached. Such conversion levels are markedly higher than the organic compounds fraction in soot. Therefore, these observations show that the presence of organic compounds in soot enhances its reactivity and permit to oxidize a carbon fraction with a higher oxidation than in the case of a soot practically free of organic compounds.

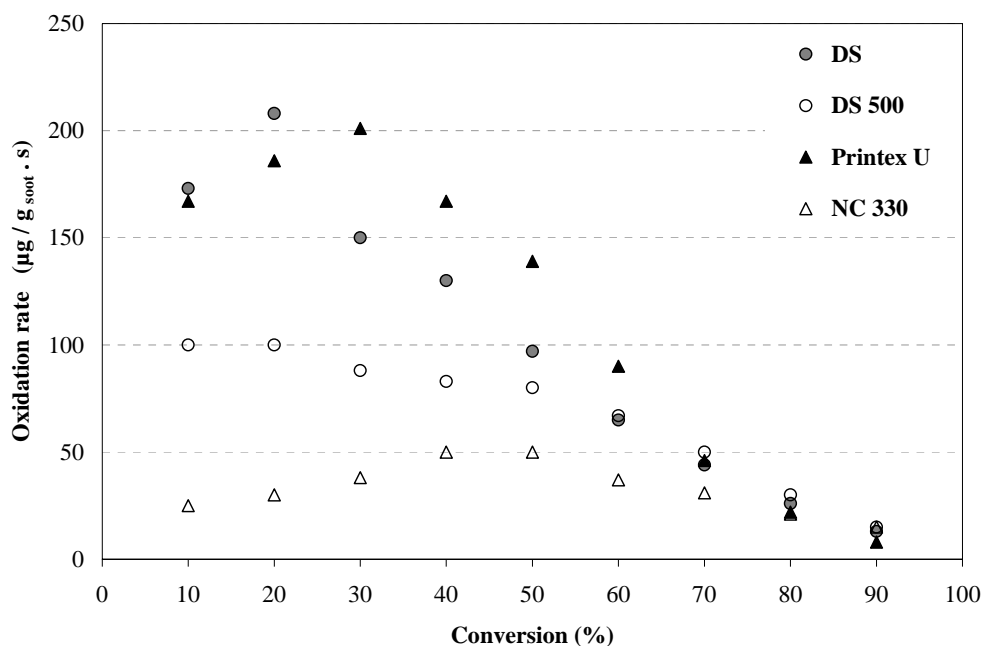


Figure 5 : Soot oxidation rate at 327°C as function of conversion in presence of $\text{Cu}_{1.96}\text{-K}_{3.45}/\text{ZrO}_2$ catalyst.

3.3. Temperature Programmed Reduction characterization of $\text{Cu-K}/\text{ZrO}_2$ catalysts

Table 3 gives the H_2 consumptions obtained for the TPR experiments of ZrO_2 , Cu_x/ZrO_2 and $\text{Cu}_x\text{-K}_y/\text{ZrO}_2$ catalysts. First, it appeared that no H_2 -consumption occurred for the ZrO_2 support while a slight H_2 -consumption was observed from 400°C for K_y/ZrO_2 . Following the copper content in Cu_x/ZrO_2 solids, the H_2 consumption increases which was ascribed to the reduction of the Cu(II) species ($\text{CuO} + \text{H}_2 \rightarrow \text{Cu}^0 + \text{H}_2\text{O}$) supported on the ZrO_2 surface. (Shimokawabe et al., 1990 ; Zhou et al. 1999). In the case of $\text{Cu}_x\text{-K}_y/\text{ZrO}_2$ samples, H_2 consumptions are markedly higher than the amounts required for the copper oxide phase reduction. Hence, it was suggested that the Cu(II) species not only interact with potassium species but also with the ZrO_2 support so that the TPR data could be explained by both the reduction of Cu(II) and species stabilized on the ZrO_2 surface. For the same Cu content (~ 2 wt% CuO), the H_2 over-consumption increases with the K content (Table 3) showing that the potassium species in the presence of copper favour the reduction of catalyst species. On the contrary, the sample with a high copper content ($\text{Cu}_{4.02}\text{-K}_{1.85}/\text{ZrO}_2$) is characterized by a relatively low H_2 over-consumption probably due to a stronger interaction of potassium species with Cu(II) than the entire solid including both Cu(II) and the ZrO_2 support.

A second step of the TPR characterization was carried out specifically to evidence if some active oxygen species of the catalysts are able to react to oxidize carbon. Such catalysts properties could account for explaining the high activity of $\text{Cu}_x\text{-K}_y/\text{ZrO}_2$. Assuming that some reducible species pointed out in the catalysts TPR profiles are active species contributing to a redox mechanism, (CB- catalysts) loose contact

mixtures previously treated *in-situ* under argon at 400°C were analyzed by TPR (see experimental part). In this case, catalysts species reduced by the carbon black during the Ar treatment could not be reduced during the subsequent H₂-TPR measurement. For solids containing only a low copper content, the H₂ consumption is unchanged. For a higher Cu content (Cu_{3,82}/ZrO₂), a decrease was obtained indicating that active oxygen species of the catalyst have reacted with carbon during the treatment of CB-catalyst mixture at 400°C. This phenomenon was ascribed to the presence of CuO crystallites considered as active species in the oxidation of carbon (Neeft et al., 1996, Pruvost et al. 2000).

Table 3: Quantitative data from TPR experiments of Cu_x-K_y/ZrO₂ catalysts.

Catalyst	Cu _{1,98} /ZrO ₂	Cu _{3,82} /ZrO ₂	Cu _{2,08} -K _{1,07} /ZrO ₂	Cu _{1,89} -K _{2,22} /ZrO ₂	Cu _{1,96} -K _{3,45} /ZrO ₂	Cu _{4,02} - K _{1,85} /ZrO ₂
H ₂ consumption (μmol H ₂ /g)	241	466	483	616	1219	679
theoretical H ₂ consumption CuO into Cu ⁰ (μmol H ₂ /g)	249	479	261	237	246	505
oxygen species released during (soot-catalyst) treatment under Ar (μmol espèces O/g)	0	131	222	379	973	174

For solids containing both potassium and copper, an important H₂ consumption decrease was recorded. These species were then reduced by carbon during the pre-treatment and this reduction is markedly high for the Cu_x-K_y/ZrO₂ with high K content. In parallel, catalytic tests performed under Ar permitted to evidence the higher conversion of carbon with higher K content in Cu_x-K_y/ZrO₂ catalysts. Our observations clearly demonstrate that potassium act as a promoter able to enhance the participation of oxygen species from the solid. Futhermore, the synergistic effects related to the addition of both Cu and K on the ZrO₂ surface could be related to the fact that potassium species facilitate the re-oxidation of active copper species involved in the catalytic reaction.

4. CONCLUSION

The characterization of diesel soot and carbon black samples show a dependence between the stabilization of particulate organic compounds and the textural properties of soots. Specific areas and porosity appears as parameters explaining the stability of organic compounds on the soot carbonaceous matrix.

The catalytic oxidation of carbonaceous samples has been investigated considering Cu and K containing ZrO₂ solids. Such catalysts are highly active systems for the oxidative elimination of soot. The organic compounds present in soot have an influence on both the soot reactivity and the catalytic reaction, as revealed by the

higher oxidation rate obtained for the Diesel soot in comparison with carbon black. Due to the presence of potassium promoter in the catalyst, the activity is related to the ability of the solid system to release active oxygen species involved in the oxidation reaction.

5. ACKNOWLEDGMENTS

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INSTRUMENTED IN-USE-VEHICLES, A VERSATILE TOOL TO MEASURE EMISSIONS

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ABSTRACT

Emission data and resulting emission factors from mobile sources provide indispensable parameters for decision making on regional air quality control.

Such data, if sourced “from other cities”, usually disqualify as being too coarse as they do not reflect the local mix of vehicles, fuel and service quality and driving behavior.

The paper describes a technology to instrument vehicles for accurate and cost- efficient acquisition of in-use motor vehicle emission data, while driving on the road. This includes a newly available on-board mobile/ portable emission measuring system, focusing on application in buses and trucks.

Until recently, such data could only be obtained with the help of laboratories equipped with complex and costly emission chassis dynos. As such facilities rarely exist for heavy-duty vehicles, these major sources of air pollution are often excluded.

Despite it’s small size and low power consumption, the new system uses laboratory-grade analyzers with inbuilt calibration gases. It delivers accurate values of emission mass as required by the legislation.

The new tool will not only close some gaps in scientific work but will also empower local authorities to better manage and verify regional air- pollution reduction undertakings.

Key Words: Real World Emissions, Emissions Factors, On Board Emissions Testing; Air Pollution Models; Mobile Emission Measurement Systems (MEMS)

1. INTRODUCTION

It is an ongoing dream of the test engineers to take measurements in-situ rather than taking the test object to a laboratory. This is even more true when it comes to the test of bulky objects such as a great number of different vehicles under the various driving conditions of a complex city.

Advances in electronics, computerization and remote positioning now make it possible to perform ”laboratory grade” measurements of mass emissions (g/km or g/h) with mobile systems operated in the driving vehicle.

This paper reports on this new solution.

2. THE TASK

Environmentalists, city and traffic planners need to know where, when and under what conditions the flowing traffic contributes to the overall pollution of a given city.

Ideally current data can be compared with future data by using alternative prediction models- some of which can be tested and evaluated at present by applying improved vehicle technology and better fuels.

THE TASK 

Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

The need and desire of cities to have a correct picture of its main transportation polluters

Three major factors influence quality:

- **Emission Factors**
Technology mix of vehicles
Age distribution of vehicles
Maintenance
Quality of fuel
- **Driving- Activity and Behavior**
- **Traffic Congestion**
Population of Vehicles


Instrumented Vehicle BAQ2004-2

3. THE TASK

As with all statistical methods the overall results improve significantly with the number of samples and the accuracy of its measurement.

There is however a trade off between cost of sampling and quality needed.

Starting with a coarse first overview followed by selected in-depth sampling usually offers optimized cost/ quality relations of good emission inventories

THE SOLUTION 

Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

- By sampling the emission of well selected vehicles during their daily cruise
- By calculating the daily/ annual total emission of each vehicle class, using statistical methods

Creating the Emission Inventory


Instrumented Vehicle BAQ2004-3

4. THE PURPOSE

Changing and improving the traffic system of a city is a cost intensive and has long lasting implications to many sectors.

It is therefore imperative for the legislator to have on hand best possible data of the presence and trustworthy predictions for the future.

To ensure sustainable improvements data validation with equal methods throughout the time line is required in addition.

THE PURPOSE 

Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

- To predict emission reductions by changing fuel quality, fuel type, applying retrofit devices or upgrading vehicle- and engine technology
- To verify predicted improvements by repeated and selected sampling after longer time intervals

Key to Policy

Instrumented Vehicle BAQ2004-4

5. THE ALTERNATIVES

For a start using emission inventories on traffic from other cities might be helpful. Inventories from cities in Industrialized countries will however provide wrong picture as technology and legislative are advanced.

Inventories from Asian cities (such as Bangkok or Shanghai) will offer more useful data in future.

Few cities have access to fully fledged emission test laboratories; even fewer have facilities allowing the testing of buses and trucks (PCD Bangkok).

This is still not much help to all "the other" cities in the region.

The modern alternative thus is to pack a "mobile emissions laboratory" to the sample vehicles- saving cost and gaining high flexibility

THE ALTERNATIVES



Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

- Using emission factors and inventories obtained by other cities or existing data bases
- Build, maintain and operate Vehicle Emission Test Laboratories
Such Labs are beyond the reach of many who need data on local emission and activity . Buses and trucks rarely covered!
- Fix Portable Emission Measuring Systems to the vehicle and obtain emission data while driving- “mobile emission laboratory“

Instrumented Vehicle BAQ2004-5

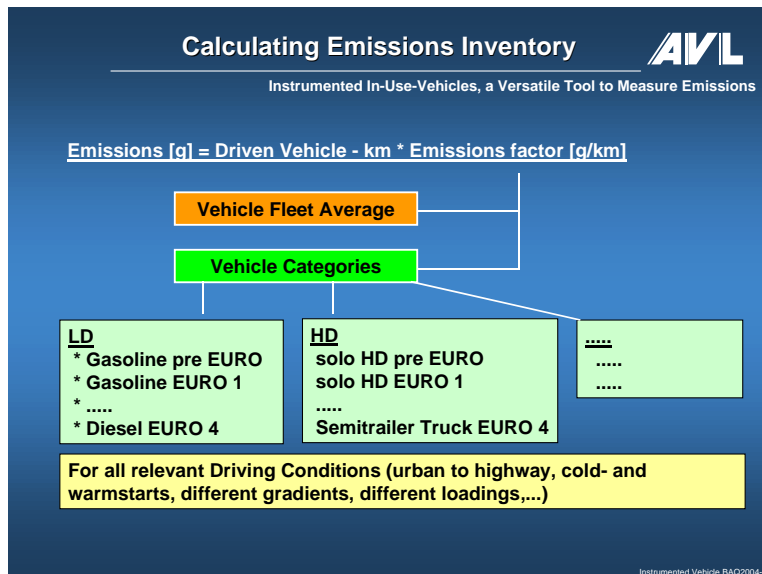
6. CALCULATING EMISSIONS INVENTORY

An emission inventory shall give the total emissions occurring in a defined area. Areas under consideration reach from single streets up to total countries. All used models have the same simple approach where the total emissions are calculated from:

with E.....total emissions in the area [e.g. g per day]
v-km.....total vehicle mileage [e.g. km per day]
e.....emission factor [g/km]

Most simulation tools used for emission monitoring are global emission and inventory models. Such models are usually based on traffic statistics and measured “emission factors” where an emission factor gives the emission value e.g. in [g/km] for a defined vehicle category in a defined traffic situation. The definition of the “average” driving cycles for a traffic situation has a high influence on the resulting emission factors. Since vehicles are driven in cold and hot running conditions, uphill and downhill, empty and full loaded in situations from congestion to free flowing traffic and with drivers having very different driving styles, a huge variety of potential using patterns exist.

As a result it is very difficult to define which driving situations are “relevant” for the air quality. Certainly all driving situations in which the most vehicle kilometer's are driven are relevant, e.g. highway driving with an average volume of traffic per lane. E.g. a total of 10 % of driving situations which have 200 % higher emission levels than the average will already give them 25 % of the total emissions (Hausberger, 2003).



7. STEPS TO MORE ACCURATE INVENTORIES

Existing Data Basis:

USA:

Mobile 5/5b, Mobile 6

Europe:

COPERT III; “German Handbook”; Artemis (not finalized)

Beside their inaccuracy the main drawback of the 2 methods shown on left in the following slide is that they can not simulate “non standard” driving cycles sufficiently. Such cycles include different gear shift behaviours, road gradients, vehicle loadings and the usage of energy intensive auxiliaries like air conditioning. All of these “non standard” situations potentially have high effects on the emission levels

Especially on HDV the influence of vehicle loading and road gradients has a very important effect.

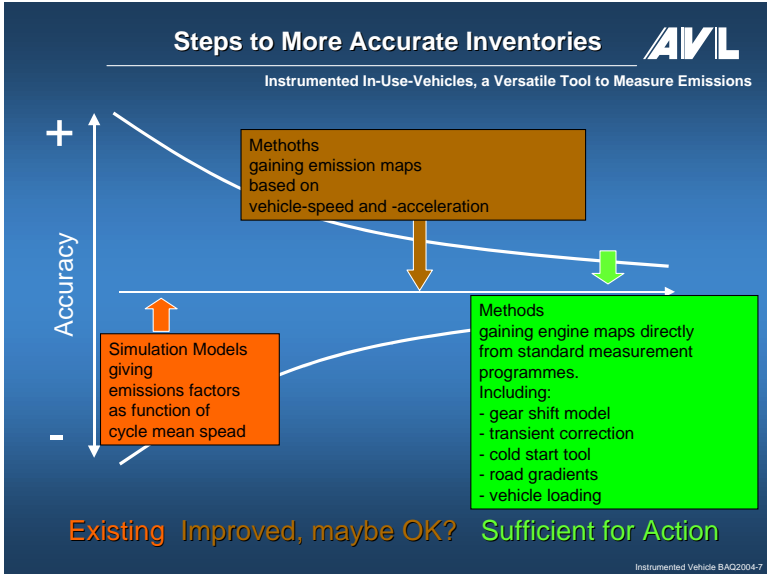
Calculated Emission Factors (g/km), at 40km/h

Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

Bus (Diesel)		"Handbook" Copert	MTC
CO	11.7	2.9	7.2
HC	3.5	1.6	2.2
NO _x	10.7	5.0	16.6
Part.	2.3	0.7	1.8

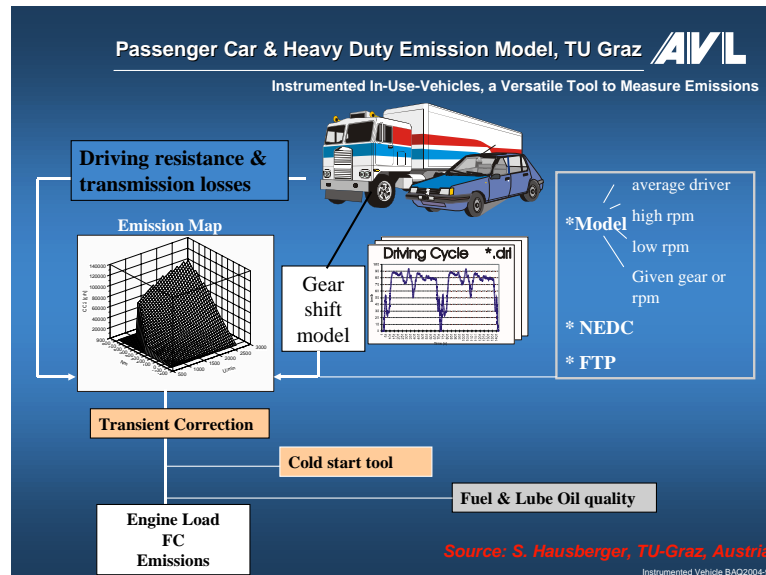
Taxi (Diesel)		"Handbook" Copert	MTC
CO	0.4	1.1	2.9
HC	0.08	0.3	0.9
NO _x	0.5	1.1	1.4
Part.	0.1	0.3	0.4

Source: L. Erlandsson, MTC Sweden
Instrumented Vehicle BAQ2004-8



8. COMPARISON OF COMMON MODELS

In the slide below Emission Factors with three different models are generated and compared.



The wealth of local factors as described affecting emissions are weighed to form the final factor

There is no single “method” to design an emission factor

Verification is essential (by the use of local emission laboratory and the help of new methods like PEMS)

9. EXAMPLE FOR A EMISSION MODEL

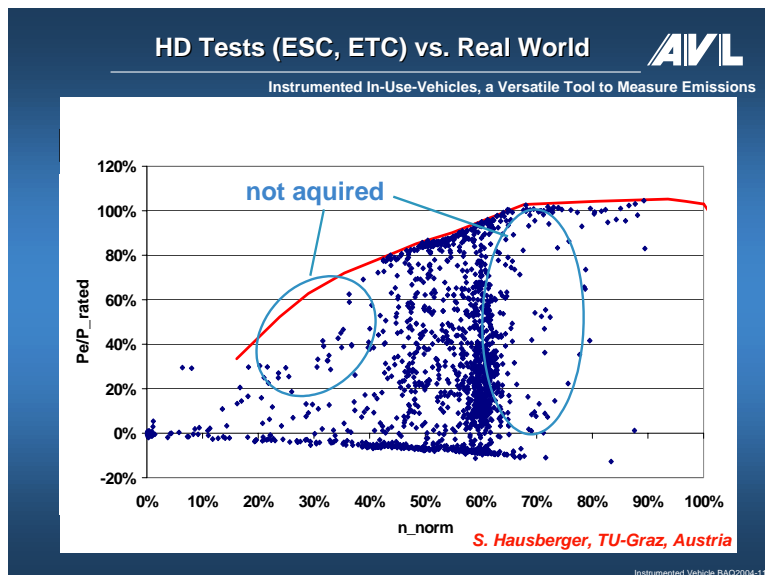
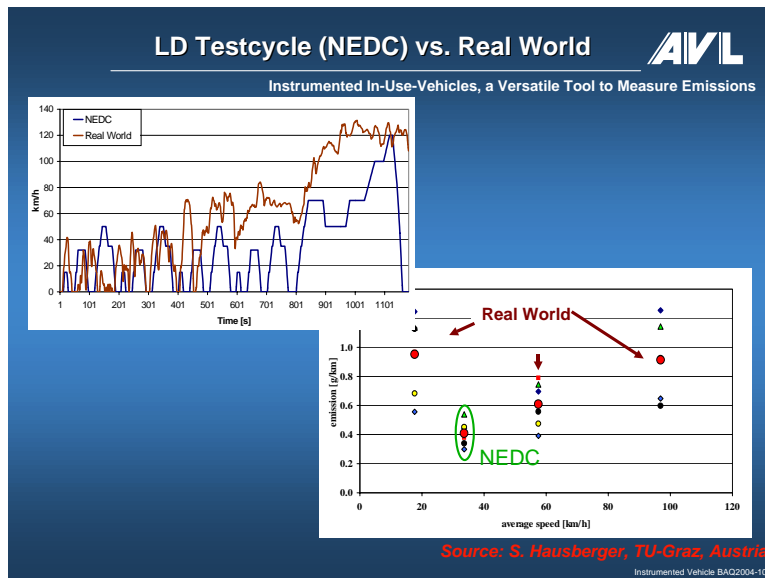
With a given driving cycle and road gradient the effective engine power is calculated in 1Hz frequency from the driving resistances and losses in the transmission system. The actual engine speed is simulated by the transmission ratios and a drivers gear shift model. The emissions are then interpolated from engine maps. Basically this method is capable of simulating the fuel consumption and the emissions for any driving cycle with any vehicle configuration.

All models described use a lot of emission measurements as model input and most of them use the data for model validation as well.

Measurements of fuel consumption and emissions are usually performed on roller test benches, engine test beds or on-board at the vehicle. Test beds have the advantage of exactly defined boundary conditions and thus a good repeatability. On-board measurements can be performed in real world traffic which allow an easy and exact recording on the road.

10. TEST CYCLE EMISSION VERSUS REAL WORLD EMISSION

The slide below shows the speed distribution of the New European Driving Cycle (NEDC), driven on a chassis dyno for Light Duty Vehicle. Additional to this legislative Cycle a “Real World Cycle”, which was recorded in real traffic, is shown. It is visible, that the real world cycle is more dynamic than the the NEDC and that the cycle mean velocity is higher. According to this the real world emissions can be higher than we expect it from the legislative measurements




The investigation of HDV emissions in real world driving behavior shows us that since the introduction of the EURO 1 limits the emission levels have not decreased in

real world driving conditions to the same extent as the emission limits for the type approval have been reduced. Main reasons are found in the more sophisticated technologies for engine control and fuel injection. On the one hand these modern technologies are a prerequisite for reducing the environmental impacts of HDV engines, on the other hand they give freedom for different specific optimizations at different regions of the engine map. Since fuel costs are a main factor for the competitiveness of HDV engines, manufacturers optimize the engines towards high fuel efficiencies wherever possible. That affects especially the NO^x emission levels. The steady state tests at the type approval can thus not ensure low emission levels for real world driving conditions. This was mainly found for EURO 2 engines tested with the R 49 steady state cycle while the European Stationary Cycle (ESC) valid for EURO 3 engines improves the situation. But still a broad range of the engine map is not controlled sufficiently.

11. REQUIREMENT FOR MOBILE EMISSION TESTING

Cause of the demand to measure and audit Real World Emissions US Environmental Protection Agency founded a initiative to develop Mobile Emission Measurement Systems (MEMS) with the following requirements.

Requirements for Mobile Emissions Testing 
Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions

US EPA Statement [G. Tierry]:

- High quality lab grade instruments
 - FID for THC (NDIR insufficient)
 - NDIR for CO
 - NDUV for NO_x (as good as chemi)
 - Microbalance for PM (coming soon)
- Full activity/environmental data collection
 - GPS, cellular modem, grade sensors, temperature, pressure, humidity, etc.
- Designed for both electronics-equipped and pre-electronics technology


Instrumented Vehicle BAQ2004-12

The contribution of transport to air pollution is of major concern in Europe. The member states of the European Union need efficient policy monitoring tools to check the in-use conformity of road vehicles with the emissions standards. The extraction of engines from heavy-duty vehicles or heavy non-road machinery to compare pollutant emissions against legislative limits is for several reasons impractical. A new approach to in-use conformity checking is needed. Therefore, the European Commission, through DG ENTR, is proposing to develop a protocol for in-use conformity checking (IUC) of heavy-duty vehicles based on the use of Portable Emissions Measuring Systems (PEMS).



A similar approach is followed by the US-Environmental Protection Agency (US-EPA) who initiated a research into the use of portable systems as a tool for in-use conformity checking of vehicles and engines. Their proposed method for IUC shall be applicable to 2007 and later model years (heavy-duty and non-road) engines.

12. APPROACH TO TEST REAL WORLD EMISSIONS

Cause of this need, AVL List GmbH of Graz and the analyzer specialist Sensors, Inc. of Saline, Michigan USA, have signed a Cooperation in august of 2004 to roll out a new tool and method of measuring “Real World Emissions”

New Approach to test Real World Emissions 

Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions



- --> Compact solution for emission analyses of all kinds of vehicles during real world driving conditions
- Simultaneously acquisition of NO, NO₂, CO, CO₂, THC and of the exhaust gas flow
- Online data acquisition of engine relevant parameters and of GPS data for calculating emissions of road trials (e.g. in g/km)

Cooperation Sensors, Inc. & AVL

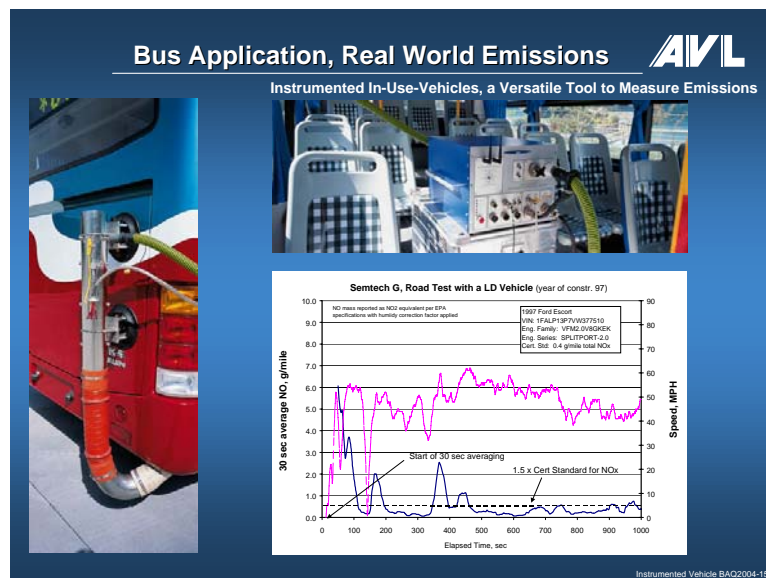
Instrumented Vehicle BAG2004-13

The SEMTECH product line was developed in response to regulatory concerns that air quality has not improved despite the increased stringency in standards over the past several decades. Deploying technology which can gather vehicle emissions information from virtually all on- and off-road vehicles makes it useful for a variety of purposes, including:

- Development of realistic mobile emissions models
- Determining the effectiveness of after treatment devices
- Meeting passenger vehicles and trucks in use compliance and NTE requirements
- Detecting and solving driveability problems
- Certification and compliance of vehicle fleets
- Supplements dynamometer testing



The diagram below shows the real world Nox emissions as a function of the driving speed. The maxima of Nox are about 4 times higher than the legislative limits for this vehicle type.





13. CONCLUSION

This paper presented a survey of methods for measuring and simulating vehicle emissions in real world driving conditions. As a result of the increased complexity of electronic engine control systems where the application has a high influence on the emission behaviour of the vehicles the established models become increasingly inaccurate.

Compared to traditional testing in emission test cells alone, on-vehicle, on-road emissions measurements using Mobile Emission Measurement Systems (MEMs) provide a broader and deeper understanding of how an engine and vehicle actually performs over its useful life. Regulatory agencies worldwide require laboratory based vehicle certification and subsequently most research to meet these requirements is carried out in emission test cells. Although remarkable improvements have been achieved this way, to make additional substantial improvements in vehicular emissions and over all air quality, it is necessary to supplement the traditional laboratory certification and research activities with in-use, on-road emission measurements using MEMs.

The accurate measurement of real world emissions levels is especially important for developing accurate emission inventories for future years and predicting effects of emission control strategies.

In-Use Vehicle Emissions Testing, Conclusions 
Instrumented In-Use-Vehicles, a Versatile Tool to Measure Emissions



- True picture of real world vehicle pollution
- Filling the gap lost by certification testing
- Efficient way of validating emissions inventories
- Possibility to measure emissions of vehicles, it does not fit into a chassis dyno

Instrumented Vehicle BAQ2004-16

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