

## An Experimental Study on Regulated and Unregulated Pollutants from a Spark Ignition Car Fuelled on Liquefied Petroleum Gas and Gasoline

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**Abstract.** In the experimental study conducted on a spark ignition (SI) car running on a chassis dynamometer, fuelled on liquefied petroleum gas (LPG) and gasoline, carbon monoxide (CO) and total hydrocarbons (HC) decreased by 37.3% and 46.8%, respectively, while oxides of nitrogen (NO<sub>x</sub>) increased by 59.7% due to higher compression ratio with LPG, compared with gasoline. In case of LPG fuel, formaldehyde, acetaldehyde, propionaldehyde, 2-butanone, butyraldehyde, benzaldehyde and valeraldehyde decreased, leading to an over all decrease of about 35% and 26% in carbonyls and their ozone forming potential (OFP), respectively, compared with gasoline. Furthermore, benzene, toluene, ethyl benzene, xylene and styrene decreased, resulting in an overall decrease of 38.8% in volatile organic compounds (VOCs) and 39.2% in BTEX (benzene, toluene, ethyl benzene and xylene) species due to more complete combustion with LPG, compared with gasoline. Further, the OFP of VOCs with LPG was 6% lower than that with gasoline fuel.

**Keywords:** liquefied petroleum gas, regulated emissions, unregulated emissions, carbonyls, gasoline car

### Introduction

During the past few years, there has been increased attention to the issues of exhausting fossil fuels deposits and increasing environmental pollution caused by vehicular exhaust, which has prompted researchers to explore alternative fuels for the transport sector. Economy, availability and environmental acceptability are the major criteria for a fuel to be accepted as an alternative fuel (Gandhidasan *et al.*, 1991). LPG is deemed to be one of the promising and viable candidates for SI engines/vehicles to resolve the issues of both rapidly depleting fuel deposits and increasing environmental concerns. It has a high octane rating and is, therefore, much suited for SI engines, but it needs additives or other positive means to initiate the combustion when it is used in compression ignition (CI) engine because of its lower cetane number (Jothi *et al.*, 2007). It is thought to be a major energy resource of the future because of its eventual availability and clean burning nature (Saleh, 2008). It is a more completely combusted fuel and hence has less impact on air quality (Chang *et al.*, 2001). LPG is widely used as an alternative vehicle fuel in the US, Canada, the Netherlands and several other countries like Japan, where 94% of the taxi fleet (about 260, 000) has been converted to LPG operation, and Australia where,

almost half a million (about 5%) of the vehicles run on LPG fuel (Ristovski *et al.*, 2005). In Hong Kong, the government has completed the scheme of the importation of LPG taxis started in 1998, before which under an incentive programme nearly all the taxis (about 99.8% of the taxi-fleet in Hong Kong) running on diesel were switched to the LPG fuel in 2003 (Ning and Chan, 2007).

LPG consists mainly of propane (nearly 80% by volume), with 11% butane and about 5% isobutane (Chang *et al.*, 2001). Due to its higher octane number, LPG can be used at higher compression ratio; consequently, the engine power and thermal efficiency (performance) are improved (Bayraktar and Durgan, 2005). It has been reported that relative to gasoline, LPG has 40% and 60% reduction in HC and CO, respectively, along with substantial reduction in carbon dioxide (CO<sub>2</sub>) emissions (Snelgrove *et al.*, 1996). According to Yang *et al.* (2007), LPG fuelled vehicles can significantly reduce the emissions of greenhouse gases, ozone forming precursors, particulate matter, metal elements, and polycyclic aromatic hydrocarbons.

The current work is aimed at the experimental investigation of CO, HC and NO<sub>x</sub> as regulated, and carbonyls and VOCs as unregulated pollutants emitted from an SI car fuelled with LPG and gasoline. In addition to this, it has been attempted to make the comparison of these pollutants in terms of their

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emission factors (EF) and corresponding OFP. Although some studies have been reported on regulated emissions from LPG-fuelled SI vehicles, unregulated emissions such as carbonyls and VOCs still need to be addressed comprehensively, particularly, in terms of their ozone formation potential under the Chinese National Standards.

Carbonyls, an important class of vehicular total hydrocarbon pollutants, play a significant role in tropospheric chemistry and are important precursors to ozone, free radicals and peroxyacetyl nitrates (Pang *et al.*, 2008). Some carbonyls are believed to be possible human carcinogens. Carbonyls such as formaldehyde, acetaldehyde and acrolein are mutagenic, toxic and even carcinogenic to human body (Carrier *et al.*, 1986); acetaldehyde and acetone have been reported to be actively involved in the photochemical smog generation cycle (Pouloupoulos *et al.*, 2001).

VOCs are thought to be major precursors of ozone, and are notorious for their health hazards and toxic properties. They play an important role in increasing ground level ozone during sunny summer periods and in the formation of secondary organic aerosols, and also contribute to the depletion of stratospheric ozone and to the enhancement of the greenhouse effect (Theloke and Friedrich, 2007). A group of VOCs consisting of benzene, toluene, ethyl benzene, and xylene isomers is known as (BTEX) pollutants. This group has the highest potentials for both ozone and peroxyacetyl nitrate (PAN) (Derwent and Jenkin, 1991). Benzene, an important component of VOCs has been declared a compound increasing the risk of leukemia (US EPA, 1990).

## Materials and Methods

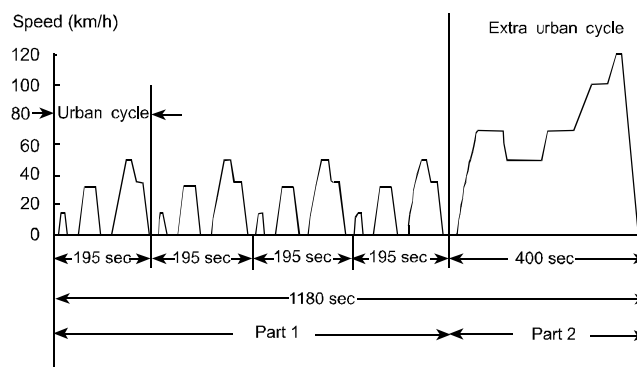
**Test vehicle, fuels, and driving cycle.** A dual-fuel 4 cylinder SI car was used in this study, which is a recent model Euro 3 compliant multi-cylinder port fuel injected (PFI) car having 1.3 L displacement volume and a maximum power of 62 kW. It was run on a 1.0 m single-roll DC electric chassis dynamometer (ONO SOKKI Inc.) following the standard protocols scheduled for the emissions of light duty vehicles (LDV). Prior to starting the experiments, the car was soaked overnight in the laboratory. In order to avoid the car from over heating, a cooling fan was used in front of it during the experiments. Two test fuels were used in this study, which are unleaded gasoline having research octane number (RON) 93 and liquefied petroleum gas (LPG), with gasoline as a reference or baseline fuel. The properties of the test fuels are listed in Table 1 (Guan-jing, 2004).

The experiments were performed in accordance with the Chinese National Standards for LDV Emissions type I test of

GB 18352.3-2005 (Yan *et al.*, 2010), which is equivalent to European cycle under directive 70/220/EEC. The cycle is shown in Fig. 1. Part one of the cycle is called urban driving cycle (780 sec) and includes four sub-cycles (195 sec each) which simulate the urban regions, and part two of the cycle is called the extra urban driving cycle (400 sec) which simulates the main motor-way, out of the urban range with more aggressive speed conditions. The total time of operation during this cycle was 1180 seconds and the average speed of the vehicle was 33.58 km/h. The schematic diagram of the experimental setup is shown in Fig. 2.

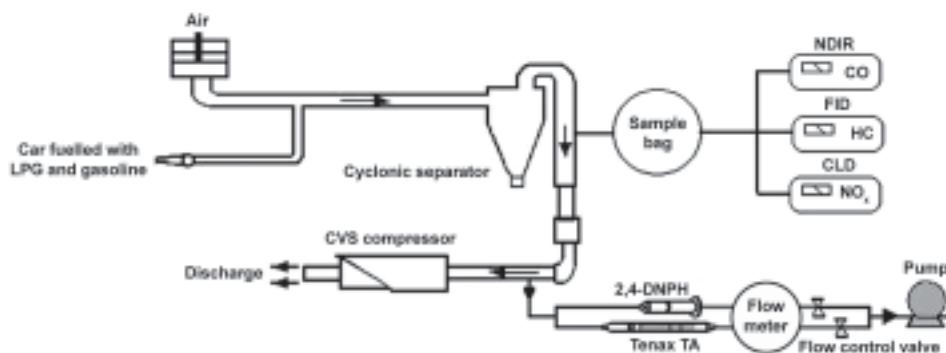
**Table 1.** Properties of test fuels (Guan-jing, 2004)

Properties	LPG	Gasoline
Density (kg/L) at 20 °C	0.582	0.74
Boiling point (°C)	-42	30-200
Auto-ignition temperature (°C)	465	450
Lower calorific value (MJ/kg)	46.4	46.0
Latent heat of vapourization (kJ/kg)	426	310
Octane number (RON)	110	93
Stoichiometric air-fuel ratio	15.3	14.7
Compression ratio	11:1	10:1



**Fig. 1.** Operating cycle for the test car in accordance with the Chinese type I test standard.

**Sampling methodology and analysis.** In order to dilute the exhaust from the car, a dilution tunnel based on standard critical flow venturi-constant volume sampler (CFV-CVS) was used. The car exhaust was introduced into the dilution tunnel where it was mixed with the fresh and filtered atmospheric air to dilute and cool the exhaust from the tailpipe, and hence eliminate the problem of water condensation during the sampling. The temperature and pressure of the diluted mixture were constantly monitored during its flow through the CFV. The dilution ratio was about 15 and the flow rate



**Fig. 2.** Experimental setup.

was  $10 \text{ m}^3/\text{min}$  in the tunnel. The dilution ratio was computed by knowing the two concentrations of  $\text{CO}_2$ . For this purpose, exhaust analyzers SEMTECH-DS and HORIBA (MEXA 7400 H) were used at the inlet and outlet of the dilution tunnel, respectively. The maximum tunnel temperature was kept below  $52 \text{ }^\circ\text{C}$  during this process. The diluted exhaust was sampled from the tunnel at two locations, one is the main location for regulated emissions and the other is at mini-tunnel for unregulated pollutants as shown in the Fig. 2.

The diluted exhaust was sampled in sampling bags (Tedlar bags) for the analysis of  $\text{NO}_x$ , CO and HC emissions. Total two bags were collected, one for the urban driving cycle and the other for the extra urban driving cycle of the complete cycle. The drive cycle emissions were collected only once in this study. Carbonyls and VOCs were sampled in 2,4-dinitrophenylhydrazine (DNPH) coated silica gel cartridges (Accustandard<sup>®</sup> Inc.) and Tenax TA<sup>®</sup> tubes (Markes USA), respectively. Each cartridge or tube was used for each transient cyclic operation. Total two tubes were used for VOCs, and five cartridges were used for the carbonyls emission analyses, then their respective average was taken over the whole test cycle. The reason for using more than two cartridges in case of carbonyls is to avoid the increased uncertainty of most of the carbonyls; particularly carbonyls such as methacrolein, valeraldehyde and aromatic aldehydes (benzaldehyde and tolualdehyde) were difficult to be identified and quantified with only two cartridges for the analysis of carbonyls. The DNPH inside the cartridges trapped the carbonyls or carbonyl compounds (CC) to react with them and to form the corresponding stable 2, 4-dinitrophenylhydrazone derivatives. The constant volume sampling pumps (SKC USA, Air Chek 2000) were used to sample the materials, and the sampling volume was kept 220 mL which took 10 min to sample at every mode. After sampling, the tubes were sealed with aluminum foil and refrigerated at  $-10 \text{ }^\circ\text{C}$ .

**Analysis of exhaust for regulated emissions.** The exhaust in the Tedlar bags was analysed for the CO, HC, and  $\text{NO}_x$  emissions. Each bag was analysed separately, and thus their average was taken in this study to discuss the pollutants. Emissions of CO in the diluted exhaust were analyzed with a non-dispersive infrared (NDIR) analyzer.  $\text{NO}_x$  were analyzed with a chemiluminescence analyzer (CLA), and total hydrocarbons were detected with a flame ionization detector (FID).

**Analysis of sampled exhaust for carbonyl emissions.** In order to extract the trapped sample material, solid phase extraction (SPE) process was used in which 2, 4-DNPH sampling cartridges were placed on solid phase extractor (USA Supelco Inc.), and sampled material was eluted from the cartridges by washing it with 3 mL acetonitrile (USA Fisher Company). The elute was collected, filtered, and then poured into a 5 mL volumetric flask to get a constant volume solution with acetonitrile. In order to elute the formed aldehydes-DNPH derivatives, a C18 column (Agilent Eclipse XDB-C18, 4.6 mm x 150 mm, 5  $\mu\text{m}$ ) was used. Acetonitrile and distilled water were used as mobile phases according to a volume ratio of 60% acetonitrile/40% water (v/v). The injected volume, flow rate and temperature gradient were 25  $\mu\text{L}$ , 1.0 mL/min and  $25 \text{ }^\circ\text{C}$ , respectively. Carbonyls were analysed using a high performance liquid chromatographic (HPLC) (USA Agilent 1200 LC) system with an automatic injector and an ultraviolet detector which detected the carbonyl-DNPHs at 360 nm. The HPLC system was used for the analysis of carbonyls.

After extraction, carbonyls were identified by matching their HPLC retention time with those of authentic standards (USA Supelco). For the quantification of the compounds, external standard method was used to make their linear standard curves. The purchased standard solutions were taken in six different concentrations with the help of a micro-sampler and were analyzed using certain chromatographic conditions to get their peak areas. According to these standard curves, the target

compounds were quantified by the regression method of their peak areas.

**Analysis of sampled exhaust for VOC-components.** For the extraction of the species trapped on Tenax TA<sup>®</sup>, automatic thermal desorber (TD) (Markes USA) was used in which Tenax tubes were first blown by the dry inert gases and then heated as discussed elsewhere (Shah *et al.*, 2009). The desorbed compounds were cryogenically focused in a cold trap at -10 °C. After focusing, the trapped material was heated from -10 °C to 280 °C at 40 °C/sec for 160 sec to volatilize the compounds into the gas chromatograph (GC) (Agilent 6890/5975) through a fused silica capillary column (HP-5MS, 30 m × 0.25 mm × 0.25 μm). Helium (99.999%) was used as a carrier gas, the oven temperature programme was from 35 °C (10 min) at 5 °C/min to 280 °C, and the column flux was 1 mL/min. The environment protection agency (EPA) standard method TO-17 and the thermal desorber-gas chromatograph-mass spectrometer (TD-GC/MS) were used to analyze the VOC species.

The VOC components were identified by comparing their retention times of chromatographic peaks with those of standard solutions purchased from Sino-Japan Friendship Center for Environment Protection, by comparing mass spectra with those available in the US National Institute of Standards and Technology (NIST05) library. After the qualitative analysis, compounds were quantified using the external standard method as discussed earlier to make their linear standard curves, and hence calibrate the curves using regression method.

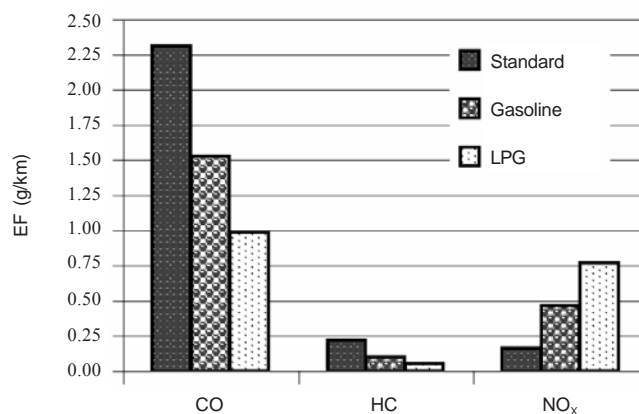
Followings were the specifications of the mass spectrometer (MS): Acquisition mode was SCAN with a range of 35-450 amu; electron multiplier voltage was 1.0 kV; transfer line to MS temperature was 250 °C; ion source was electron impact (EI) 70 eV with a temperature of 200 °C; and solvent cut time was 2.5 min.

## Results and Discussion

**Regulated emissions.** In this study, both the regulated and unregulated emissions have been discussed in terms of their emission factor which is defined as the mass of the pollutants emitted from the car per kilometer of distance traveled by it. The reason to discuss emissions in terms of EF is to facilitate a comparison between different combinations of cars/vehicles and the test fuels.

As presented in Fig. 3, carbon monoxide and total hydrocarbon emissions from both the fuels were well below the standard value. Relative to gasoline, LPG showed 37.3% and 46.8% decrease in CO and HC emissions, respectively. This

decrease in CO and HC emissions is attributed to increase in combustion efficiency of the car engine when running on LPG. Since, LPG has a higher octane number compared to gasoline, which permits a higher engine compression ratio to be used for LPG operation, which improves combustion efficiency and engine power with increased cylinder temperature. Higher temperature in the cylinder plays an important role in promoting the rate of oxidation of the fuel (Cheung *et al.*, 2008), and hence may reduce the CO and HC emissions. Moreover, LPG has a higher air/fuel ratio relative to gasoline, which improves the peak flame temperature, rate of flame propagation, and oxidation rate, and thus lean-burn LPG engine has lower emissions of CO and HC. In addition, the higher hydrogen/carbon ratio of LPG is principally responsible for CO and HC reduction (Snelgrove *et al.*, 1996). According to Rice *et al.* (1991), a significant decrease in pollutant emissions may be achieved by operating the gasoline engines on fuel-lean mixtures and/or at higher compression ratio. Gaffney and Marley (2009) have reported that vehicles which use gaseous fuels can reduce CO emissions because they can run on stoichiometric air/fuel ratio during cold start, when CO pollutants are at their peaks.



**Fig. 3.** Comparison between gasoline and LPG fuels on the basis of emissions regulated under the type I test standard.

As shown in Fig. 3, oxides of nitrogen are produced from both the fuels in higher quantity as compared to their standard values. This indicates that NO<sub>x</sub> emanated from both the fuels do not meet the Chinese state standard type I test for light duty vehicles. As far as the comparison of NO<sub>x</sub> between the two fuels is concerned, LPG exhibits higher emissions as compared to gasoline fuel. Relative to gasoline, NO<sub>x</sub> emissions increase by 59.7% in case of LPG fuel. This increase in NO<sub>x</sub> emissions is ascribed to the higher combustion temperature

in case of LPG, compared with gasoline fuel. This finding is consistent with the study of Yang *et al.* (2007).

**Unregulated emissions. Carbonyl emissions and their ozone forming potential.** As shown in Table 2, acrolein and acetone have been discussed together in this study because it was difficult to separate them in the column due to their almost identical retention time. Experimental results show that in case of LPG formaldehyde, acetaldehyde, propionaldehyde, 2-butanone, butyraldehyde, benzaldehyde and valeraldehyde decrease by 66%, 56.5%, 38.8%, 63.4%, 57.1%, 76.3% and 38.1% respectively, while acrolein+acetone, methacrolein and hexanaldehyde emissions increase by 41.8%, 71.4% and 65.2% respectively, relative to gasoline. As a result, there is an over all decrease of 34.7% in carbonyl emissions with LPG, compared with gasoline fuel.

**Table 2.** Comparison between LPG and gasoline fuels on the basis of carbonyl emission (mg/km) and the corresponding SR (number of replicates = 5)

Carbonyls	LPG	Gasoline	MIR	(SR) <sub>LPG</sub>	(SR) <sub>Gasoline</sub>
Formaldehyde	0.82±0.374	2.41±0.243	7.15	0.87	1.72
Acetaldehyde	1.37±1.442	3.15±0.410	5.52	1.12	0.75
Acrolein+acetone	3.02±0.865	2.13±0.427	0.56*	0.25	0.12
Propionaldehyde	0.41±0.212	0.67±0.261	6.53	0.40	0.63
2-Butanone	0.34±0.286	0.93±0.212	1.18	0.06	0.11
Methacrolein	0.12±0.125	0.07±0.086	6.77	0.12	0.05
Butyraldehyde	0.06±0.104	0.14±0.042	5.26	0.05	0.13
Benzaldehyde	0.09±0.102	0.38±0.154	-0.56	-0.01	-0.03
Valeraldehyde	0.13±0.191	0.21±0.112	4.41	0.09	0.09
Hexanol	0.38±0.218	0.23±0.105	3.79	0.21	0.42
Σ	6.74	10.32		3.16	4.27

0.56\* is for acetone.

The decrease in the emissions of carbonyl compounds in case of LPG is due to the difference in physico-chemical properties of the two fuels. Properties like higher octane number, higher flame temperature, faster flame propagation speed, higher compression ratio, and lean-burn nature of LPG advance the combustion process when it burns in the engine cylinder. According to Magnusson *et al.* (2002), formaldehyde from the vehicle exhaust results from the incomplete combustion of saturated aliphatic hydrocarbons which are frequently found in gasoline. So, LPG fuel results in more complete combustion, relative to gasoline. The reduction in benzaldehyde with LPG is quite understandable, which is due to the less aromatic and olefin contents of LPG as compared to gasoline. However, the increase in acrolein+acetone, methacrolein and hexanaldehyde with LPG may be due to the restraint in the oxidation of their precursors, compared with gasoline.

Formaldehyde, acetaldehyde, acrolein+acetone, and benzaldehyde contribute 78.6% and 78.1% of the total carbonyls emitted with LPG and gasoline fuels, respectively. This finding is in good agreement with those of previous studies that formaldehyde, acetaldehyde, acrolein+acetone, and aromatic aldehydes (benzaldehyde and tolualdehyde) contribute to maximum emissions of total carbonyls with gasoline fueled spark ignition engines (Pang *et al.*, 2008; Magnusson *et al.*, 2002).

The ozone forming potential of the pollutants can be calculated on the basis of their specific reactivity (SR) which is defined as the mass of ozone (O<sub>3</sub>) potential (in mg) per milligram mass of the non-methane organic gas (NMOG) for the emissions, and can be calculated using the following relation (Lowi and Carter, 1990).

$$SR = \frac{\sum(NMOG_j \cdot MIR_j)}{\sum NMOG_j} \quad (1)$$

where:

subscript j stands for a certain compound other than methane or non-methane organic gas (NMOG) like carbonyls and VOCs and

MIR stands for maximum incremental reactivity.

Lowi and Carter (1990), examined air modelling based on ozone forming reactivity of species and proposed the MIR factor as an index for ozone formation, which was issued by the California Air Resource Board (CARB) in 1992.

Table 2 shows the comparative SR of emissions from the test fuels, evaluated on the basis of MIR values. The results show that specific reactivity of carbonyls emissions in case of LPG is 8.5% lower, compared with gasoline. This finding is understandable, and is due to the lower carbonyls emissions, particularly the decrease in formaldehyde emissions with LPG, compared with gasoline fuel.

**VOCs emissions and their ozone forming potential.** As presented in Table 3, benzene, toluene, ethyl benzene, p-m-xylene, styrene and o-xylene decrease by 25.9%, 40.6%, 39.7%, 39.4%, 7.5% and 32.9%, respectively, while butyl acetate and n-undecane emissions increase by 48% and 24% respectively, in case of LPG compared with gasoline. Consequently, there is an overall decrease of 34.2% in VOCs emissions with LPG fuel, relative to gasoline. The BTEX pollutants decrease by 39.2% in case of LPG, compared with gasoline fuel.

The reason for the reduction of VOCs, particularly BTEX emissions is the more complete combustion, and hence higher combustion temperature in case of LPG, relative to gasoline.

**Table 3.** Comparison between LPG and gasoline fuels on the basis of VOCs (mg/km) and their corresponding SR (n = 2)

VOCs	LPG	Gasoline	MIR	(SR) <sub>LPG</sub>	(SR) <sub>Gasoline</sub>
Benzene	0.20	0.27	0.42	0.002	0.001
Toluene	23.77	40.01	2.73	1.31	1.45
Butyl acetate	5.21	3.52	n/a	n/a	n/a
Ethyl benzene	5.02	8.32	2.7	0.27	0.30
p,m-Xylene	7.19	11.87	7.64	1.11	1.21
Styrene	0.86	0.93	2.22	0.04	0.03
o-Xylene	6.54	9.75	6.46	0.85	0.84
n-Undecane	0.67	0.54	0.42	0.01	0.003
Σ	49.46	75.21		3.60	3.83

Higher temperature is prone to decompose the VOCs emissions; particularly benzene, toluene, and xylene isomers decrease significantly at higher temperature (Di *et al.*, 2009; Cheung *et al.*, 2008). Similar findings have been reported by Gaffney and Marley (2009) that total VOC components reduce with CNG and LPG when compared with gasoline or alcohol fuels. Furthermore, LPG contains less aromatic content and has a higher hydrogen/carbon ratio, both of which are responsible for the reduction of VOC species, compared with gasoline (Yang *et al.*, 2007). On the other hand, the reason for the higher emissions of butyl acetate and *n*-undecane with LPG, relative to gasoline may be the release of their uncombusted precursors, particularly from the cars operating without catalytic converters.

As far as their OFP is concerned, VOCs from LPG exhibit 6% lower SR, relative to gasoline as shown in Table 3. The above finding is in accordance with the literature that the atmospheric reactivity of the organic emissions from LPG decreases appreciably as compared to gasoline (Gaffney and Marley, 2009). This reduction in SR of VOCs in case of LPG, compared with gasoline is due to the reduction in the emission of VOCs, particularly xylene and toluene carrying the highest MIR values among the VOC-components.

### Conclusion

In the current study, it has experimentally been attempted to examine the effect of LPG on the regulated and unregulated pollutant emissions of an SI car. It was found that the regulated emissions such as CO and HC were decreased by 37.3% and 46.8% respectively, while the NO<sub>x</sub> pollutants increased by 59.7% with LPG, compared with gasoline fuel. This might be due to the increase in compression ratio of SI engine with LPG relative to gasoline, due to which both CO and HC emissions have decreased but NO<sub>x</sub> increased. Although the

NO<sub>x</sub> were higher, while the CO and HC pollutants were well below the type I test standard values with both the test fuels. Furthermore, most of the carbonyls such as formaldehyde, acetaldehyde, propionaldehyde, 2-butanone, butyraldehyde, benzaldehyde and valeraldehyde were decreased mainly due to the higher octane number, higher flame temperature and higher compression ratio with LPG, compared with gasoline. In addition, the OFP of carbonyls with LPG was 25.7% as lower as with gasoline. Moreover, VOCs emissions such as benzene, toluene, ethyl benzene, p,m-xylene, styrene and o-xylene were highly reduced with LPG due to more complete combustion, relative to gasoline fuel. In case of LPG, an overall decrease of 38.8% in VOCs and 39.2% in BTEX species were revealed compared with gasoline. Further, the contribution of VOCs to OFP was 6% lower with LPG, relative to gasoline fuel.

### Recommendation

The simultaneous reduction in all three major pollutants i.e., in CO, HC and NO<sub>x</sub> can possibly be achieved with LPG by decreasing the compression ratio of the car engine under LPG operation. This action, no doubt, will increase the HC and CO emissions. However, all the three pollutants can be reduced to their optimum possible values through this way.

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