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Continuous effectiveness of replacing catalytic converters on liquified petroleum gas-fueled vehicles in Hong Kong

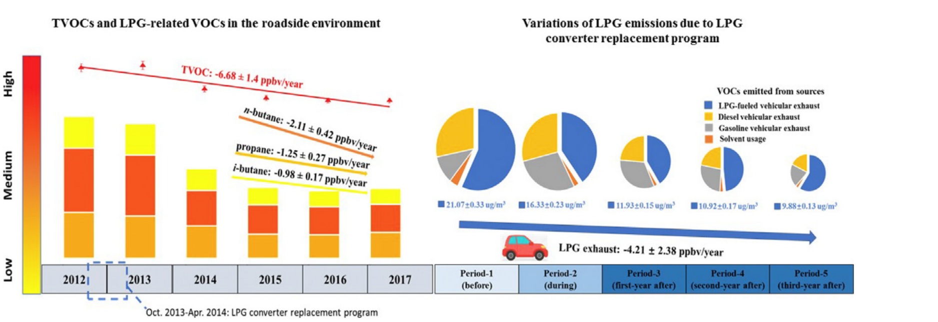
Dawen Yao [a], Xiaopu Lyu [a], Frank Murray [b], Lidia Morawska [c], Wang Yu [a], Jiaying Wang [a], Hai Guo [a,*]

[a] Air Quality Studies, Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong
[b] School of Environmental Sciences, Murdoch University, Perth, Australia
[c] International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia

HIGHLIGHTS

- VOCs emitted from LPG-fueled vehicles decreased
- Net negative contribution of VOCs and NO_x from LPG vehicles to O_3 increased
- The intervention program has been continuously effective.

GRAPHICAL ABSTRACT



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ABSTRACT

To mitigate the concentrations of air pollutants in the atmosphere, an intervention program of replacing the converters of liquefied petroleum gas (LPG) fueled vehicles was implemented by the Hong Kong government between October 2013 and April 2014. Data of ambient volatile organic compounds (VOCs) and other trace gases continuously monitored from September 2012 to April 2017 at a roadside site were used to evaluate the continuous effectiveness of the replaced catalytic converters on the reduction of air pollutants. The measurement data showed that LPG-related VOCs (propane and *n*/*i*-butanes) and several trace gases (CO, NO and NO_2) decreased significantly from before to after the program ($p < 0.01$). To further assess the efficiency of the program, five periods covering before the program, during the program, 1st year after the program, 2nd year after the program and 3rd year after the program were categorized. The values of propane and *n*/*i*-butanes decreased from Period-1 (before the program) to Period-2 (during the program), and from Period-2 to Periods 3–5 (after the program) ($p < 0.01$). In addition, the reduction rates of propane and *n*/*i*-butanes remained high and constant in Periods 3–5, suggesting that either had the vehicle owners themselves routinely replaced the converters at suitable interval afterwards, or were their vehicles caught by a remote sensing program checking excessive emissions. Source apportionment analysis indicated that LPG-fueled vehicular emissions were the top contributor to ambient VOCs in the roadside environment while the VOCs emitted from LPG-fueled vehicles indeed decreased at a rate of 4.21 ± 2.38 ppbv/year (average ± 95% confidence interval) from Period-1 to Period-5 ($p < 0.01$). Furthermore, the photochemical box model simulations revealed that the net negative contribution of VOCs and NO_x emitted from LPG-fueled vehicles to O_3 production strengthened at a rate of 1.9×10^{-2} pptv/day from Period-1 to Period-5 ($p < 0.01$). The findings proved the continuous effectiveness of the intervention program, and are of help to future control strategies in Hong Kong.

* Corresponding author.
E-mail address: ceguohai@polyu.edu.hk (H. Guo).

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1. Introduction

During the past decades, many cities in the world including Hong Kong have been suffering from severe ozone (O_3) pollution (Jacob, 1999; NARSTO, 2000; So and Wang, 2003; Lam et al., 2005; Wang et al., 2017, 2018). It is essential to mitigate O_3 pollution due to its adverse effects on the environment and human health (Sillman, 1999; Seinfeld and Pandis, 2006). Being key O_3 precursors, volatile organic compounds (VOCs) and nitrogen oxides (NO_x) are important chemicals contributing to high O_3 production in Hong Kong and the adjacent Pearl River Delta (PRD) region (Zhang et al., 2007; Shao et al., 2009; Guo et al., 2007, 2013; Wang et al., 2017). Previous studies revealed that O_3 formation is sensitive to VOCs in urban areas of this region (Cheng et al., 2010; Ling et al., 2011), and subsequently it is a foreseeably effective way to control O_3 pollution through constraining the VOCs concentrations in the atmosphere (Ling et al., 2011). Hence, understanding the characteristics of VOCs and quantifying their source apportionments are fundamental for the formulation and implementation of O_3 control strategies.

To improve air quality, the Hong Kong government has implemented a series of control measures in the past decades, mainly focusing on the reduction of air pollutants from emission sources such as motor vehicles and solvent usage (HKEPD, 2007, 2014). With the execution of these control measures, VOCs emissions from some sources have been under control (Ling and Guo, 2014; Huang et al., 2015; Lyu et al., 2017). For example, Huang et al. (2015) found that VOCs emitted from diesel and gasoline vehicles decreased by at least 37% in 2011 compared to those in 2003 at the same roadside site in Hong Kong. Lyu et al. (2017) reported that VOCs emitted from diesel exhaust and solvent usage decreased whereas the gasoline and liquefied petroleum gas (LPG) vehicle emissions elevated from 2005 to 2013. Indeed, due to the widespread LPG use in public transport since 2001, VOCs emissions from LPG-fueled vehicles and their contributions to O_3 production gradually increased in the last decade (Guo et al., 2011; Ho et al., 2013; Ou et al., 2015; Lyu et al., 2016a, 2017). Specifically, LPG usage even accounted for 30% of the ambient non-methane hydrocarbons (NMHCs) in suburban Hong Kong (Ou et al., 2015). With the aid of the positive matrix factorization (PMF) model, Lyu et al. (2016a) revealed that LPG fuel consumption was the largest source of VOCs at a roadside site during October 2012–September 2013, accounting for 50% of the total VOCs in Hong Kong.

To specifically reduce air pollutant emissions from LPG-fueled vehicles in Hong Kong, an intervention program of replacing the catalytic converters on LPG-fueled vehicles was undertaken from October 2013 to April 2014 (HKEPD, 2014). In this program, a total of about 17,000 taxis and light buses took part in (Environment Bureau, 2017). To assess the immediate effectiveness of this program on pollutant emissions from LPG-fueled vehicles, Lyu et al. (2016a) compared the data at a roadside site before the program (June–September 2013) with that after the program (October 2013–May 2014), and claimed that VOCs and nitrogen oxides (NO_x) emitted from LPG-fueled vehicles significantly reduced, notwithstanding very minor increase of O_3 production (Lyu et al., 2016a). Despite instant usefulness, it was reported that the catalyzing capacity of the converters would gradually fade away with the time of use (Heck and Farrauto, 2001; Sharma et al., 2014). Since the mileages of many LPG-fueled vehicles are very high (e.g. several hundred kilometers per day for many taxis), the catalytic converters need to be replaced approximately every 18 months (Simpson et al., 2015). Hence, it is necessary to evaluate the continuous effectiveness of the intervention program on the reduction of air pollutant emissions from LPG-fueled vehicles.

The main objective of this study was to assess the continuous effectiveness of replacing catalytic converters on LPG-fueled vehicles in Hong Kong. To achieve it, the evolution of air quality at a roadside station in Hong Kong was studied between 2012 and 2017, a long period covering the three stages of before, during and after the implementation of the replacement program. The long term trends of LPG tracers were examined, and a detailed comparison was conducted among different stages of this program. Furthermore, a PMF model and a photochemical box model incorporating Master Chemical Mechanism (PBM-MCM) were applied to understand the changes of the contribution of LPG-fueled vehicles to ambient VOCs and the ground-level O_3 production caused by the replacement program.

2. Methodology

2.1. Sampling site

Continuous measurements of target air pollutants were conducted at an urban roadside location in Hong Kong, namely Mong Kok (MK) site. MK site (22.32°N, 114.17°E, 3 m a.g.l.) is one of the three urban roadside monitoring stations (MK, Causeway Bay and Central) set up by the Hong Kong Environmental Protection Department (HKEPD). The site is located at the junction of Nathan Road and Lai Chi Kok Road, Mong Kok, Kowloon (Fig. 1), which is surrounded by residential and commercial building blocks with heavy daily traffic (Lee et al., 2002). As an urban roadside station, MK site is less influenced by regional air masses and the main local sources are traffic emissions (Lee et al., 2002; Cui et al., 2016).

2.2. Measurements of target air pollutants

2.2.1. Trace gases

Trace gases including SO_2, CO, O_3, NO and NO_2 were continuously monitored at MK from September 2012 to April 2017. CO was analyzed using the method of non-dispersive infrared absorption with gas filter correlation; SO_2 was detected with UV fluorescence analyzers; NO–NO_2–NO_x was measured by chemiluminescence technique; An UV photometric analyzer was used to monitor O_3 mixing ratio. The detection limits for CO, SO_2, NO, NO_2 and O_3 were 50.0, 1.0, 0.5, 0.5 and 2.0 ppbv, respectively. All the instruments were regularly calibrated and tested with the instruments and quality control and assurance (QC/QA) procedures identical to those in the US air quality monitoring program (Ou et al., 2015; Lyu et al., 2016a; Wang et al., 2017).

2.2.2. Volatile organic compounds (VOCs)

Non-methane hydrocarbons (NMHCs) were measured by an online analyzer (Syntech Spectras GC 955, Series 600/800, Netherlands). The instrument is an integrated separating and analytical system consisting of two sampling systems and two columns for separation of the C_2-C_5 hydrocarbons and the C_6-C_{10} hydrocarbons, respectively (HKPU, 2012). The target species included 30 VOCs (10 alkanes, 10 alkenes, 1 alkyne and 9 aromatics). Ambient air samples were collected and analyzed every 30-minute continuously. The 30-minute concentrations were averaged into hourly values for further analysis (Ou et al., 2015; Lyu et al., 2016a; Wang et al., 2017).

2.3. Quality assurance and quality control (QA/QC)

Strict QA/QC procedures were followed to assure the data quality. Built-in computerized programs of QC system, e.g., auto-linearization and auto-calibration were developed for the analyzer. Weekly calibrations were conducted by injecting certified calibration gas (National Physical Laboratory span gas). Additionally, independent comparisons with canister samples analyzed by UC-Irvine were carried out. Overall, the detection limits of the target NMHCs ranged from 2 to 787 pptv. The accuracy of the measurements was about 1–10% and the measurement precision was about 2.5–20%. More details about the QA/QC protocols can be found in the previous studies (Xue et al., 2014; Ou et al., 2015; Lyu et al., 2016a).