



Refueling emissions from cars in Japan: Compositions, temperature dependence and effect of vapor liquefied collection system



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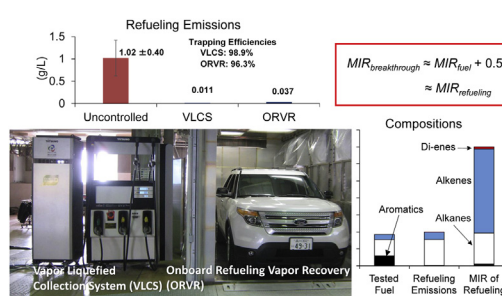
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HIGHLIGHTS

- Refueling emission factor in Japan was 1.02 ± 0.40 g/L and MIR was 3.49 ± 0.83 .
- C₄ alkene had the highest impact on the OFP of refueling emissions.
- Refueling emissions changed with temperature but MIR was almost constant.
- $MIR_{\text{refueling}} \approx MIR_{\text{breakthrough}} \approx MIR_{\text{fuel}} + 0.5$.
- The efficiency of a vapor liquefied collection system was similar to that of an ORVR.

GRAPHICAL ABSTRACT



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ABSTRACT

Refueling emissions from cars available on the Japanese market, which were not equipped with specific controlling devices, were investigated. For the composition analysis, a proton transfer reaction plus switchable reagent ion mass spectrometry (PTR + SRI-MS), which is capable of real-time measurement, was used. In addition, the performance of a vapor liquefied collection system (VLCS), which is a recently developed controlling device, was evaluated and compared with an onboard refueling vapor recovery (ORVR) system. The refueling emission factor of uncontrolled vehicles at 20 °C was 1.02 ± 0.40 g/L in the case dispensing 20 L of fuel. The results of composition analysis indicated that the maximum incremental reactivity (MIR) of refueling emissions in Japan was 3.49 ± 0.83 . The emissions consist of 80% alkanes and 20% alkenes, and aromatics and di-enes were negligible. C₄ alkene had the highest impact on the MIR of refueling emissions. The amounts of refueling emissions were well reproduced by a function developed by MOVE2010 in the temperature range of 5–35 °C. The compositions of the refueling emissions varied in this temperature range, but the change in MIR was negligible. The trapping efficiency of VLCS was the same level as that of the ORVR (over 95%). The MIRs of refueling and evaporative emissions were strongly affected by that of the test fuel. This study and our previous study indicated that $MIR_{\text{breakthrough}} \approx MIR_{\text{refueling}} \approx MIR_{\text{fuel}} + 0.5$ and $MIR_{\text{permeation}} \approx MIR_{\text{fuel}}$. The real-world estimated average MIR_{fuel} in Japan was about 3.0.

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1. Introduction

Efforts have been made in many countries to reduce emissions of volatile organic compounds (VOCs) to improve the ozone

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situation. The emissions from automobiles are thought to be one of the major sources of VOCs. It is known that the VOCs from automobiles are emitted not only from the tailpipe during driving but also from the fuel tank of gasoline vehicles during parking and refueling. These are called evaporative emissions and refueling emissions, respectively. These emissions have been studied since the 1980's, mainly in the USA (Braddock et al., 1986; Braddock, 1987; Berglund and Petersson, 1990; Furey and Perry, 1991; Burns et al., 1992). For the evaporative emissions, recent studies have paid attention to estimating the amount in real-world conditions. Our previous study (Yamada, 2013) showed that the total evaporative emissions in Japan are higher than the tailpipe emissions, accounting for 4.6% of VOC emissions from stationary sources. In Europe, the evaporative emissions in real-world conditions often exceed the regulation limits (Martini et al., 2014). Dong et al. (2015) proposed an updated method for estimating the emissions from real parking activity data more accurately. On the other hand, there have been hardly any recent studies on refueling emissions. This is probably because prevention techniques have been already established and introduced in many countries.

The strategy for preventing refueling emissions in the USA is to employ an onboard refueling vapor recovery (ORVR) system which traps the vapor displaced by the liquid fuel entering into the tank by using a carbon canister installed in the car. Unlike the USA, in Europe, the refueling emissions are vacuumed by a stage II system installed on a gasoline dispenser (European Parliament, 2009). These two are the major techniques in use, and usually countries which wish to control refueling emissions choose one of them after considering their merits and demerits. The ORVR system exhibits high trapping efficiency (95%). However, the benefits of ORVR will only be fully delivered after most vehicles on the road are replaced with those having ORVR, and this may take more than 10 years. In contrast, although the real world trapping efficiency of stage II systems is only 70%, they can be implemented more quickly (Fung and Maxwell, 2011). The theoretical efficiency of stage II system is similar to that of ORVR; in reality, however, they are often not well-maintained, which results in lowered efficiency. Japan is one of the countries where refueling emissions have not been controlled, although its ozone situation is critical (Ministry of Environment, Japan, 2011). In Japan, gas stations emitted 107,082 ton/year out of the total VOC emissions of 736,612 ton/year (Ministry of Environment, Japan, 2014). Therefore, the introduction of prevention techniques has been demanded by local governments in Japan (Kanagawa Prefectural Government, 2014), and discussions will begin in the near future. However, there is no data on the effectiveness of control techniques for refueling emissions in Japan. As described above, there were many studies discussing the effectiveness of these techniques in the USA in the 1980's. However, the current situation in Japan is quite different from the situation in the USA in the 1980's, such as car specifications, fuel composition, and fuel dispenser specifications. The results of these evaluations will be useful not only to Japan but also to many other countries that have not yet introduced refueling emission controls.

To discuss VOC emissions from various sources, it is important to consider the ozone formation potential (OFP) (Adam et al., 2011; Zhang et al., 2013; Gentner et al., 2013; Costagliola et al., 2014; Li et al., 2015), but there have been hardly any studies that discussed the OFP of refueling emissions in Japan. Even in the studies performed outside Japan, in most cases, the composition analyses were made using traditional analytical chemistry methods, such as GC-MS. Our previous study demonstrated that measurement of evaporative emissions using proton transfer reaction plus switchable reagent ion mass spectrometry (PTR + SRI-MS) can be performed in real-time. Also, the results indicated the possibility of underestimating the concentrations of relatively high-molecular-

weight compounds due to adsorption on the walls of the variable-temperature sealed housing evaporative determination (VT-SHED) equipment (Yamada et al., 2015). Therefore, real-time measurements of VOCs are desirable in evaporative emission experiments.

The Japanese gasoline dispenser maker Tatsuno has developed a new prevention device that can be deployed by replacing existing dispensers with new dispensers equipped with the device, even though there have been no regulations concerning refueling emissions in Japan. Their approach uses a vapor liquefied collection system (VLCS). Similar to the stage II system, the refueling emissions are vacuumed by a muffler-shaped inlet surrounding the nozzle. VLCS may be an effective tool; however, no evaluation of its emission preventing performance has been carried out.

In this study, the refueling emissions from eight Japanese vehicles not equipped with refueling emission control systems were measured using a VT-SHED and a gasoline dispenser. In these experiments, the maximum incremental reactivity (MIR; Carter, 2010; Carter and Heo, 2013), which is one of the major indicators of OFP, was estimated from the results of real-time composition analysis with the PTR + SRI-MS. Changes in the amounts of refueling emissions and their compositions were also observed in the temperature range from 5 to 35 °C. In addition, the performance of the VLCS was compared with that of the ORVR.

2. Experimental methods

2.1. Refueling emissions measurements

The refueling emissions from gasoline vehicles were measured by placing a test vehicle in a VT-SHED (VSH-9353; Hitachi Technology Engineering Inc., Tokyo, Japan) that met the requirements for the Japanese approval tests for evaporative emissions but that was not designed for refueling emission measurements. The VOC emissions were determined by measuring the increase in the total hydrocarbon (THC) concentration in the SHED using a gas analyzer (MEXA-1160TFL-L; Horiba Inc., Kyoto, Japan).

A gasoline dispenser (MAB36621EVMBDTX0001A; Tatsuno Corporation, Tokyo, Japan) was set next to the SHED and the dispenser nozzle was brought into the SHED through a square-shaped hole with approximate dimensions of 150 mm × 150 mm just before the test started. The hole was loosely sealed with a polytetrafluoroethylene sheet. Any leak from the hole was considered to be negligible because the hole was small enough compared with the SHED volume of 93.53 m³ and because of the relatively short test durations of approximately 5 min. The test was started upon removing the fuel tank cap and then inserting the nozzle of the dispenser into the tank as quickly as possible. Initially almost 10% of the fuel was left in the tank, and 20 L of gasoline was filled into the tank. The gasoline flow rate was set to 35 L/min, which is the rate generally used in Japan. After refueling, the nozzle was left inserted, and the test ended when the indicated THC stabilized. The test fuel was specially blended for type approval tests, and its temperature was set equal to the SHED temperature. This procedure roughly agrees with that in the US. The differences are, in the US case, background measurement is performed with the tank cap opened and fuel is dispensed until automatic nozzle shut-off occurs (US Government Publishing Office, 1995). The shape of the nozzle was almost the same as the stage II nozzle which is surrounded with a muffler-shaped vacuum inlet. The vacuumed flow was sent to the VLCS (QE-1001; Tatsuno Corporation, Tokyo, Japan) at a flow rate of 40 L/min. The same nozzle was used even when the VLCS was off, for conducting measurements of the ORVR and the uncontrolled case.

Alkanes, alkenes and di-ene species concentrations from carbon

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