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Evaporative emissions in three-day diurnal breathing loss tests on passenger cars for the Japanese market



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HIGHLIGHTS

• 4 out of 7 Japanese vehicles exhibited breakthrough emissions in 3-day DBL tests.

• Evaporative emissions based on test results are estimated to be 32,792 t/y in Japan.

• Real-time measurements showed adsorption of HC onto the SHED during DBL tests.

• MIRs of breakthrough and permeation emissions were 3.89 and 3.31, respectively.

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ABSTRACT

Breakthrough emissions that dominate diurnal evaporative emissions from gasoline vehicles were observed in continuous 3-day diurnal breathing loss (DBL) tests. These measurements were conducted on nine vehicles for the Japanese market. Two of these vehicles, made by US and European manufacturers, also meet regulations in their countries of origin. Four vehicles exhibited marked emissions caused by breakthrough emissions during the experimental period, all made by Japanese manufacturers. Using our experimental results, we estimate the total diurnal evaporative emissions from gasoline vehicles in Japan to be 32,792 t y⁻¹. The compositions of the breakthrough and permeation emissions were analyzed in real time using proton transfer reaction plus switchable reagent ion mass spectrometry to estimate the ozone formation potential for the evaporative emissions. The real-time measurements showed that the adsorption of hydrocarbons in a sealed housing evaporative determination unit can result in underestimation, when concentrations are only monitored before and after a DBL test. The composition analysis gave an estimated maximum incremental reactivity (MIR) 20% higher for the breakthrough emissions than for the gasoline that was tested, while the MIR for the permeation emissions was almost the same as the MIR for the fuel. Evaporative emissions from gasoline vehicles in Japan were found to contribute 4.2% to emissions from stationary sources using a mass-based estimate, or 6.1% of emissions from stationary sources using a MIR-based estimate.

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1. Introduction

High concentrations of photochemical ozone in the urban atmosphere are a problem, even in developed countries. Elevated ozone concentration episodes occur occasionally in California (US Environmental Protection Agency, 2012) and around the Mediterranean Sea (European Environment Agency, 2013). Japan has a serious problem with photochemical ozone, only six of the 1183 ozone concentration measurements that were made in 2010 were

* Corresponding author. E-mail address: h-yamada@ntsel.go.jp (H. Yamada). below the national standard (Ministry of the Environment Japan, 2011). Ozone is produced in the atmosphere through reactions between volatile organic compounds (VOCs) and NO_x (Milford et al., 1994). Therefore, it is important to decrease emissions of these compounds, so that less ozone is formed and air quality is improved.

Automobiles are major sources of VOCs and NO_x. NO_x is emitted from the tailpipes of automobiles, only while running; while VOCs are emitted by gasoline vehicles from their tailpipes, and as evaporative emissions. Evaporative emissions have been well-studied in the USA (Furey and Perry, 1991; Pierson et al., 1999; Rubin et al., 2006; Gentner et al., 2009) and have been regulated since the





ATMOSPHERIC

1970's. Current regulations in the USA are the most strict worldwide; they require a running loss test, given that this is considered to be one of the major VOC emission sources from light duty gasoline vehicles (Kirchstetter et al., 1996; Lough et al., 2005), as well as a 2- or 3-day diurnal breathing loss (DBL) and hot soak loss (HSL) tests. Regulations in Europe and Japan still only require 1 day DBL and HSL tests, which were phased out in the USA decades ago.

There may be attempts to improve air quality in Japan and Europe by introducing stricter evaporative emission regulations, similar to those in place in the USA. Investigations into updating evaporative emission regulations were recently undertaken in Europe (Martini et al., 2007, 2014). In Japan, there are few studies that discuss the VOC inventory in Japan (Chatani et al., 2011; Kurokawa et al., 2013). However, only our previous study addresses evaporative emissions in Japan (Yamada, 2013a). In that study, the total amount of diurnal evaporative emissions consisting of "permeation" (the leakage of fuel from the fuel tank and piping) and "breakthrough" (the overflow of fuel vapor from the carbon canister) were estimated. Evaporative emissions were mainly dominated by the breakthrough emissions, which were found to contribute 4.6% of the total emissions of VOCs from stationary sources in Japan. However, these estimates are speculative, based on a single experiment and meteorological data for Tokyo. Breakthrough emissions vary with car models because their fuel tank designs and carbon canister volumes are different. In addition, diurnal evaporative emissions are affected by meteorological conditions. Hence, data showing their variation with different car models and different regional weather conditions are needed to improve the estimation of total evaporative emissions. A great deal of such data has been produced by researchers in the USA, but all involving cars for the US market. As stated above, regulations for evaporative emissions are quite different between Japan and the USA, including the techniques that are used to prevent such emissions (Manufacturers of Emission Controls Association, 2014). Thus, it is important to study the evaporative emissions from gasoline vehicles for the Japanese market.

It is well known that different VOC species have different impacts on the formation of ozone. Therefore, it is important to assess the effects of VOC emissions using an ozone formation potential (OFP) parameter (Dimitriades, 1996; Kirchstetter et al., 1999; Martien et al., 2003; Chen and Luo, 2012), such as maximum incremental reactivity (MIR) (Carter, 2010; Carter and Heo, 2013). Hence, evaluations of automobile exhausts using an OFP have often been performed not only on tailpipe exhaust (Yang and Milford, 1996; Olson et al., 2000; Schmitz et al., 2000; Farrugia and Olson, 2005; Nakashima et al., 2010; Adam et al., 2011; Tsai et al., 2012; Wang et al., 2013; Costagliola et al., 2014), but also on evaporative emissions (Burns et al., 1992; Harley et al., 2000; Gentner et al., 2013). In those studies, the VOC compositions were analyzed using traditional analytical chemistry techniques, such as gas chromatography with flame ionization detection or gas chromatographymass spectrometry (GC-MS). Such techniques are excellent methods for identifying and quantifying VOC species, but samples of automobile exhaust gas for analysis have to be stored in Teflon bags or canisters, resulting in loss of hydrocarbons with relatively high molecular weights (Zielinska et al., 1996). Such losses will also be a rather serious issue for DBL tests using a sealed housing evaporative determination (SHED) unit, because such tests last for 24 h, with measurements usually only conducted before and after a test.

In this study, we conducted 3-day DBL tests on nine vehicles that have been used in the Japanese market. We selected these vehicles to outline the variability of breakthrough features among different car models. Two of these vehicles were made by US and European manufacturers; hence, they meet the Japanese regulation, as well as the regulations in their country of origin. We provide an estimate of the total diurnal evaporative emissions from gasoline vehicles in Japan based on these 3-day DBL test results, taking into consideration regional meteorological data. We also estimated the OFPs for permeation and breakthrough emissions produced during the DBL tests using on-line VOC data acquired using proton transfer reaction plus switchable reagent ion mass spectrometry (PTR + SRI-MS) (Jordan et al., 2009).

2. Experimental methods

2.1. Evaporative emissions measurements

The evaporative emissions from gasoline vehicles were measured by placing a test vehicle in a variable temperature SHED (VSH-9353; Hitachi Technology Engineering Inc., Tokyo, Japan) that met the requirements for the Japanese approval tests for evaporative emissions. Each test lasted for 24 h. The temperature inside the SHED was raised from 20 to 35 °C and then reduced again to 20 °C, such that the temperature profile reproduced the diurnal changes on a sunny day; this is standard procedure for a DBL test. We performed each DBL test three consecutive times to reproduce the evaporative emissions produced by a vehicle on three consecutive days. 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). To prepare for a test, the test vehicle was driven for more than 1 h at 80 km h^{-1} to clear the adsorbed fuel from the carbon canister. Gasoline, specially blended to meet the requirements of the Japanese approval tests, was then added to the fuel tank, filling the tank to 40% capacity. The test vehicle was then left in the SHED for more than 8 h at 20 °C, which was the initial temperature used in each DBL test. During this soak period of the DBL test, the ventilation system of the SHED was used to avoid increasing background THC. THC concentrations were confirmed not to exceed 3 ppmC before each test. After each test, the ventilation system was also used to avoid contamination of the SHED from emissions, until THC concentration dropped below 3 ppmC.

We tested nine vehicles that are in current use in Japan (Table 1). Seven of these vehicles were made by Japanese manufacturers, one was made by a European manufacturer (vehicle FC), and one by a US manufacturer (vehicle FM). All of the vehicles met the requirements of the Japanese regulations, however the evaporative emission control devices in vehicles FC and FM were similar to those used in their manufacturers' native markets, given that the regulations in those markets are similar to or stricter than the Japanese regulations. All cars had previously traveled between 7600 and 168,000 km and were between 0.8 and 13.6 years old. The evaporative VOC emissions from vehicle CB were analyzed in detail by PTR + SRI-MS.

2.2. PTR + SRI-MS analysis

Alkanes, alkenes, and aromatic hydrocarbons are major components of gasoline (Erickson et al., 2014). Recently, we reported that alkane species can be ionized using the switchable reagent ion (SRI) reaction (Eq. (1)) using NO⁺ as the reagent ion (Inomata et al., 2014).

$$NO^+ + RH \rightarrow R^+ + HNO$$
 (1)

Alkanes can, therefore, be monitored by analyzing the ion produced in the reaction shown in Eq. (1), which has an m/z ratio of the molecular weight -1. This allowed the composition of the evaporative emissions from vehicle CB to be measured using Download English Version:

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