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How ethanol and gasoline formula changes evaporative emissions of the vehicles

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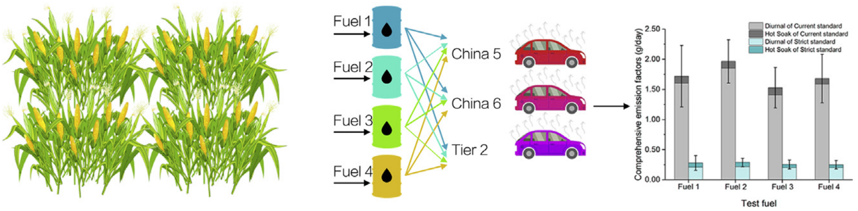
HIGHLIGHTS

- Study aims at the largest vehicle market and the significant emission process.
- Fills the gap on understanding evaporative emission impacts from ethanol.
- New fuel formula, new vehicle technologies were tested.
- Largest scale evaporative emission tests in recent years.
- A comprehensive model and regulation evaluation for China.

GRAPHICAL ABSTRACT



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ABSTRACT

China, the biggest vehicle market in the world, will implement nation-wide use of ethanol-added gasoline (contains 10% ethanol, E10) by 2020. This change will have significant impact on evaporative emissions which contribute 40% of total vehicular volatile organic compounds (VOCs) in China. This study performs the largest scale measurements on vehicle evaporation in China utilizing four types of market-based gasoline (three E10 gasolines versus one E0 gasoline) and 5 vehicles with two control levels: normal control (major fleet in Euro and China) and advanced control (US Tier 2 and future China 6). Add of thanol and aromatics components enhance emissions through permeation mechanism, while Reid Vapor Pressure (RVP) has more impacts on canister-venting emissions. Average hot soak emissions increased by 45.4%, 40.5% and 28.6% when using E10 fuels for Euro 4, China 6 prototype and Tier 2 vehicles compared to emissions using E0 fuel. The average permeation of Euro 4 vehicles increased by ~60% compared emissions when using E0 fuel. While, the impacts on diurnal emissions are associated with the dominated emission mechanism. Considering the real-world control strategy, the best performance on evaporative emissions was achieved by Fuel 3 (E10 with low RVP and aromatic). Besides, an upgrade on emission standard could dramatically reduce the total emission amount and the effects due to fuel composition difference can be ignored, indicating using ethanol would not become an excuse for violating compliance with emission regulations including China 6, which would be implemented in China in 2020.

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1. Introduction

Ethanol has been utilized as a gasoline blend stock in the U.S. since the early 1980s and promoted national with passage of several acts e.g. the Clean Air Act Amendments (CAAA) of 1990, the Energy Policy Act (EPAct) of 2005 and the Energy Independence and Security Act (EISA) of 2014. Essentially all U.S. gasoline contains 10% ethanol (E10), with the ethanol component being produced nearly exclusively from corn starch [1]. Sugarcane has been used as raw material to produce ethanol in Brazil [2], and at least 27% percentage of ethanol should be added into gasoline now [3]. Other Raw materials saccharins groups, such as sugar beet, sweet sorghum, sweet potato est., are also used in many countries [4–6]. Future production of renewable transportation fuels such as ethanol must rely on abundant nonfood plant sources also known as lignocellulosic biomass [7–10]. Right now, the usage of fuel ethanol was limited to 11 provinces in China and the consumption of fuel ethanol accounts for merely 0.8% of the total petroleum products [11]. However, China will implement nation-wide use of ethanol-added gasoline (E10) by 2020, while targeting the large-scale production of cellulose ethanol and advanced biofuel technologies by 2025 [12]. It's the first time the government has set a timeline for promoting the biofuel, known as E10, referring to gasoline with an ethanol content of 10 per cent. Considering that China is the largest vehicle market in the world, the change in vehicle emissions that would result from a national large-scale conversion from gasoline to E10 could have significant environment impacts.

Now, China has been suffering serious air pollution especially $PM_{2.5}$ and ozone, and vehicle emissions have been an important contributor to air pollution in Chinese cities [13–15]. Secondary organic aerosol (SOA) accounts for a significant fraction of ambient tropospheric aerosol [16,17]. Gas-phase oxidation of VOCs has traditionally been considered to be an important source of urban SOA formation [18,19]. Studies on ozone pollution also demonstrated that ozone formation was controlled by VOCs in many major Chinese cities [20,21].Vehicle-related emissions are widely recognized as the main source of anthropogenic volatile organic compounds (VOCs) in urban areas, contributing more than 50% of the annual ambient VOC concentrations [15,22,23]. VOC emissions from motor vehicles are generated through several pathways that can be grouped into tailpipe emissions (or exhaust) and non-tailpipe (or evaporative) emissions. Compared with vehicular tailpipe emissions, vehicular evaporation emissions has been proved to be a non-ignorable contributor to the ambient VOCs concentrations recently [24–26]. Lab measurements showed that the evaporative emission factor of one Euro4/5 gasoline vehicle is 0.21 g/km [27], much higher than the Euro3 tailpipe emissions (0.12 g/km). From the national vehicle fleets perspective, the vehicular evaporative VOC emissions in China are 1.65 Tg in 2015, accounting for 39.20% of total vehicular VOC emissions [24]. In addition, with stricter exhaust control regulations, the portion of evaporative emissions to total VOCs emissions is increasing contrary to the downward trend on tailpipe emissions.

So the first major consideration of promoting ethanol blends is the impacts of vehicle emissions. There are plenty of emission experiments on E10 gasoline. While, most of the studies focus on tailpipe exhausts not evaporation. These studies reported that using ethanol blended gasoline can reduce the exhaust emissions of carbon monoxide (CO) and hydrocarbon (HC) by 5.3–61% depending on the content of ethanol [28–37]. According to a summary of existing studies on impacts of ethanol (Table 1) [29,38–45], relevant researches are very scarce especially in China. Even the handful evaporation studies get controversial conclusions. For hot soak, testing results obtained by Paz et al. indicated a 12% decreasing achieved by E5 and E10 fuels comparing to test results based on E0 fuel [38]; while other hot soak tests conducted on two Japanese vehicles (model year 2000) by Tanaka et al. found a 2.6 and 6 times increasing using E3 fuels comparing to emissions using E0 fuel [39]. What's more, Delgado et al. found no significant influence on hot soak emission when conducted tests using E5, E10 and E85 ethanol blends. For permeation and diurnal emissions, the results are also confusing. A fuel permeation project reported by CRC (Coordinating Research Council) stated that E6, E10, and E20 ethanol blends significantly increased the permeation based on tests conducted on ten Californian in-fleet vehicles [45]. Martini et al. conducted diurnal tests on four gasoline vehicles with European 6 Standard, showing an increasing trend after using ethanol blends [40]. While, some analysis also reports a reduction from 1.05 g/day to 0.87 g/day after using E10 fuels on 24-h diurnal tests [42].

We noticed that most of the test fuel ethanol used in previous studies was made up by adding ethanol directly to base E0 fuel (regular reformulated gasoline) (Table 1). The compositions of gasoline in different countries are different due to the weather, oil supplier and other conditions. Thus, a proven mechanism of ethanol blending is: adding ethanol directly into gasoline will increase the RVP in the system, and will reach the highest point when the ethanol concentration is between 2% and 10% [46]. Increment of RVP will enhance fuel vapor generating and evaporative emissions [47]. In reality, instead of directly adding a specific percentage of ethanol into regular gasoline (laboratory experiment procedure), percentages of other organic compounds, such as aromatics, alkanes and alkenes, will also be adjusted based on economic efficiency in gasoline reformulating process. Such difference will also cause divergent influence on evaporative emission. Therefore, conclusions drawn from research using laboratory allocated fuel ethanol will deviate from the real-world situation. It is necessary that ethanol blended gasoline formulated during researches is consistent with the future real-world situation e.g. the actual portion of productions; additional researches based on this criterion should be done to seek a more precise prediction.

Another matter lies in whether ethanol becomes an excuse for violating emission standards. Among evaporative emission regulations, U.S. and Euro standards are both specified that the emission limits are also applied to E5 ethanol fuels during emission tests [48,49]. Currently, emission standards in China are based on non-ethanol gasoline [50]. If 10% ethanol does impact emissions, will it make vehicles change from attainment to non-attainment? This question becomes very sensitive for industry. Meanwhile, China's new emission standard (China 6), which reduces evaporative emission standards from 2 g/day to 0.7 g/day, is about to be effective in 2020 [51]. Comparing to European and U.S. regulatory requirements, China 6 has a pre-conditioning for hot soak testing at high temperature (38 ± 2 °C) following WLTC (Worldwide harmonized Light vehicles Test Cycles) and a 12–36 h high-temperature soak (38 ± 2 °C) before a high-temperature driving test. The last two test procedures only appeared in China 6 so far. It's already a challenge to automobile industry. Will ethanol gasoline make a harder time for them? No previous study can answer this question. Simulating an application of ethanol blends on current Chinese automobile fleet along with the upgraded automobile fleet could provide an insightful depiction of future fuel policy.

In this study, 41 crossover tests rank as one of the top largest evaporation measurements in the world. Distinguished from laboratory single-variable experiments, this study provides insights for three options for future ethanol blended gasoline formulas and compared them with current gasoline formula. The evaporative emissions, including hot soak emissions, 24–48 h diurnal emissions and permeations, were measured using orthogonal experiment for three types of evaporation control technology vehicles and four types of emission measurement procedures. The results were then combined with the China vehicle fleet and previous investigation results to build a national model. The comprehensive impacts from possible E10 formula were evaluated based on China vehicle fleet. The critical components for controlling evaporation from E10 gasoline were also investigated to find out the mitigation solution. The sensitive issue about failing to meet the emission standards was touched and some quantitive results were provided.