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Exposure to hazardous volatile pollutants back diffusing from automobile exhaust systems

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HIGHLIGHTS

- ► The emission of hazardous volatiles was investigated from back diffusion gases of automobiles.
- ► Pollutant emission patterns vary greatly between the different driving conditions.
- ► Back diffusion can be a significant source of in-vehicle pollution to passengers.

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ABSTRACT

As back diffusion gases from automobiles are significant sources of in-vehicular pollution, we investigated eight automobiles, five for back diffusion (driving) measurements and three for reference conditions (non-driving). To characterize the back diffusion emission conditions, seven volatile organic compounds (VOC) and four carbonyl compounds (CCs) were measured along with dilution-to-threshold (D/T) ratio. The data obtained from back diffusion measurements were examined after having been divided into three subcategories: (i) driving and non-driving, (ii) with and without automobile upgrading (sealing the inner line), and (iii) differences in CO emission levels. Among the VOCs, the concentrations of toluene (T) was found to be the highest (range: 13.6–155 ppb), while benzene (0.19–1.47 ppb) was hardly distinguishable from its ambient levels. Other VOCs (xylene, trimethylbenzene, and styrene) were generally below <1 ppb. Unlike VOCs, the concentrations (ppb) of CCs were seen at fairly enhanced levels: 30.1−95 (formaldehyde), 34.6−87.2 (acetaldehyde), 4.56−34.7 (propionaldehyde), and 3.45−68.8 (butyraldehyde). The results of our study suggest that the back diffusion phenomenon, if occurring, can deteriorate in-vehicle air, especially with the most imminent health hazards from a compound such as formaldehyde in view of its exceedance pattern over common guidelines.

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1. Introduction

An automobile is defined as four wheeled motor vehicle powered by an internal engine and used for transporting passengers [1]. Around the world, about 1 billion cars and light trucks were on the road in 2010, most of which were driven by gasoline [2]. In the case of South Korea, there has been a 130-fold increase in the total number of automobiles from 1970 to 2008, amounting to 16.79 million units [3].

Several previous investigations attributed mobile sources like vehicle exhaust as the dominant source of hazardous volatile organic compounds such as benzene, toluene, xylenes, and so on in ambient air [4–9]. Like VOCs, carbonyl compounds (CC) are also identified as a major hazardous pollutant group emitted from motor vehicle exhaust [10–12]. As such, mobile sources can account for the emission of such key volatile pollutants as benzene, acetaldehyde, and formaldehyde, by 60, 39, and 33%, respectively [13].

Automobile exhaust gas mixtures contain minute carbon particles (<1 μm) that may be inhaled into the lungs along with hazardous compounds [14]. Like adults, unborn children can also be affected if pregnant women are exposed to high concentrations of toluene [15]. Although the acute effects of VOCs have commonly been observed with a number of symptoms (such as asthma), long-term exposure can lead to an increased risk of cancer or damage to the liver, kidney, and central nervous system [16].

Unlike VOCs, concentration levels of aldehydes, especially acetaldehyde and propionaldehyde from back diffusion gases are highly sensitive components according to the malodor regulation guidelines [3]. In addition, formaldehyde and acetaldehyde are also listed as probable carcinogens [17]. Long-term exposure of formaldehyde and acetaldehyde can lead to an increased risk of cancer. It was found that professional drivers are prone to a high cancer risk [18,19]. By damaging red blood cells (RBC),

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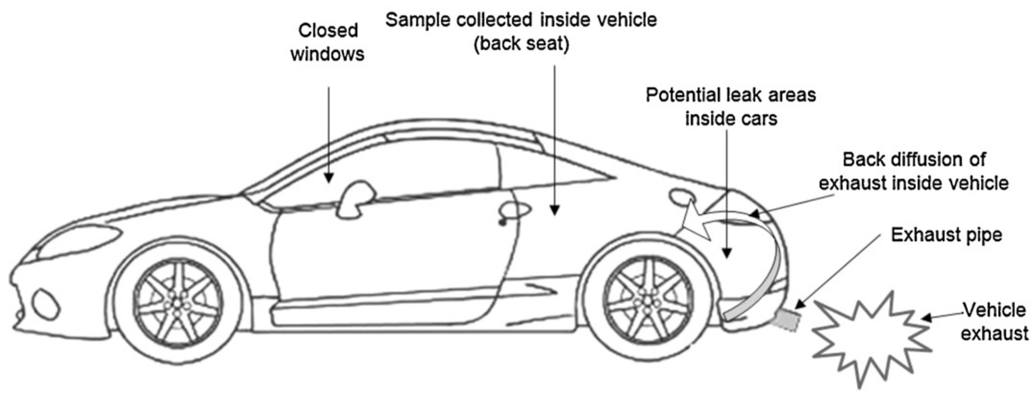

Fig. 1. Schematic vehicle sampling site, exhaust flow at rear of the vehicle, and potential sites for exhaust back diffusion into the rear passenger area.

acetaldehyde can modify hemoglobin (oxygen transporter), which can then cause greater oxygen deficiency in cells, especially in the brain [20]. To protect the human population from adverse health effects, the New Zealand Ministry of Environment set up guidelines for ambient acetaldehyde concentrations (as annual average) at 16.9 ppb [21]. Formaldehyde, if its concentration exceeds 100 ppb, is also known to cause irritation of the eyes, nose, and throat. As such, it is capable of accelerating asthma symptoms and other respiratory illnesses [22–24]. Moreover, in some cases, it was reported that acute exposure (230 ppm) of butyraldehyde (BA) can cause coronary injury [25].

Mobile emissions of VOCs are of increasing concern due to their involvement in the photochemical smog and adverse health impact via inhalation exposure at certain levels [26,27]. Consequently, the U.S. Environmental Protection Agency set up environmental regulations and standards to induce the reduction of exhaust emissions of non-methane hydrocarbon (NMHC), e.g., for new gasoline-fueled passenger vehicles, 0.3 g/miles for light ($\leq$6000 lbs) and 0.5 g/miles for heavy (>6000–10,000 lbs) vehicles [28].

In this research, we studied in-vehicle exposure of VOCs and CCs from samples collected inside eight different automobiles. As exhausted gases can have a hazardous impact on commuters, we measured their concentration levels diffused back (Fig. 1) into automobiles under varying conditions. The phenomenon of back diffusion can be defined as exhaust gas penetrating inside the automobile through the backside leaking parts of vehicle instead of going out through exhaust pipe (Fig. 1). The concentration levels of the back diffusion gases were measured inside automobiles and evaluated in relation to a number of variables under driving conditions (e.g., automobile upgrading (sealing back diffusion routes), driving speed, and varying levels of CO). As the main criteria to assess the back diffusion conditions, the concentrations of VOC and CCs were also analyzed from inside automobiles under non-driving reference conditions in car parking areas.

2. Materials and methods

2.1. Sample collection

The selection of the testing vehicles was made to include the Grandure III 2011 model, Hyundai motor company (purchased individually by local dealers). As certain models of this vehicle have been reported to have back diffusion problems, this single model vehicle with gasoline (octane) fuel was used entirely for our investigation. To this end, a total of eight target automobiles were selected voluntarily by the owners who claimed to suffer from the back diffusion phenomenon to a certain extent (Table 1). To diagnose the status of back diffusion from these eight target automobiles, a total of 19 samples were collected to simulate both the back diffusion (driving) and reference (parking) conditions.

For the collection of samples under back diffusion conditions, vehicles selected in this study were driven on the highway in the outskirts of Seoul. Detailed information concerning the disposition of test vehicles is summarized in Table 2. During sampling, windows were closed to reduce the diffusion of outside air (or emissions from outside the vehicles). Three passengers (one for driving and two for sampling in the back seat position) were seated in each model vehicle for the tests throughout the study. To analyze automobile pollution due to back diffusion, gaseous samples inside the vehicles were collected in the back seat position in eight automobiles. Among these eight automobiles, five were investigated to collect samples under the back diffusion conditions, while three were used as reference conditions. During sample collection for reference conditions, the vehicles were parked in the car parking areas with their engines stopped. To discover the emission trends of VOCs, a total of 19 samples were collected, which can be categorized into two data groups, reference ($n = 6$) and back diffusion cases ($n = 13$). All 13 samples of the latter case were further divided into three sub-categories: (1) right after engine starting vs. high speed driving (at speed of 100–120 km h^{-1}), (2) before upgrading (with back diffusion evident) vs. after upgrading (after adding sealant in the backside leaking areas), and (3) between different CO levels (above and below 10 ppm) (Table 2). Collected gas samples were analyzed for the listed VOCs and CCs in Table 1.

For the analysis of the main target pollutants of VOCs and the D/T ratio, samples were collected using 10 L Polyester Aluminum (PEA) bags with the aid of a lung vacuum sampler (ACEN Co. Ltd., Korea). The duration of bag sample collection varied slightly with regards to the status of the vacuum build-up conditions of the samplers at each sampling. However, the collection of each bag sample was generally completed in less than 10 min. In addition, for the analysis of CCs, samples were also drawn into Lp-DNPH cartridges (Supelco Inc., US) at a flow rate of 1 L min^{-1} (for 5 min) via a Sep-Pak ozone scrubber (Waters, US).

2.2. Analysis of target compounds

All collected samples were analyzed to determine concentrations of up to 21 volatile pollutant species (Table 1). As a simple means to assist the diagnosis of back diffusion, the extent of odor possibly generated from exhaust gas was also measured in terms of the D/T ratio by the air dilution sensory (ADS) test [29]. The basic analytical settings and experimental conditions for the analysis of automobile air pollutants inside vehicles are described in Table 3. The detailed analytical procedures of these target pollutants have been described in a number of our previous investigations [30–32].

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