<table>
<thead>
<tr>
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<tr>
<td><strong>Author(s)</strong></td>
<td>Imamura, Kiyoshi; Maeda, Yasuaki; Takenaka, Norimichi; Pham, Hung Viet; Tran, Thi Ngoc Lan</td>
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</table>

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Osaka University
INVESTIGATION ON AIR POLLUTION IN VIETNAM
-VOLATILE ORGANIC COMPOUNDS IN HANOI AND HO CHI MINH-

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4) Vietnam National University, Hanoi, Vietnam
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ABSTRACT
The ambient levels of 40 saturated and unsaturated aliphatic hydrocarbons, 10 aromatic hydrocarbons, and 16 halogenated hydrocarbons were determined in Hanoi and Ho Chi Minh City. Relative standard deviations of ambient levels for all aliphatic and aromatic hydrocarbons were more than 80%, because of the contribution of their different sampling sites from the emission source. An ambient level of isopentane, toluene, and m,p-xylene was high in both cities. The levels of halogenated hydrocarbons (trichlorofluoromethane (freon 11), dichlorodifluoromethane (freon 12), 1,1,2-trichloro-1,2,2-trifluoroethane (freon 113), and carbon tetrachloride) were low and almost uniformed at all sampling sites. Chloromethane and dichloromethane utilized as solvents were the abundant components, and their levels were low in both cities.

Constituents of four types of gasoline available in the market were determined for comparison, and contribution ratios of gasoline to atmospheric pollution levels were estimated to be 3-57% and 8-56%, and those of LP&Exhaust to be 8-40% and 10-32% for Hanoi and Ho Chi Minh City respectively.

KEYWORDS
aliphatic, aromatic, and halogenated hydrocarbons, concentration level of components, contribution ratio, gasoline components, Hanoi and Ho Chi Minh City, urban atmosphere

INTRODUCTION
The volatile organic compounds (VOC) have been widely used as the fuels of liquefied petroleum (LF), gasoline, kerosene, and the industrial materials including the solvents for painting. Some of them (Exhaust) are exhausted from the combustion process and the cracking thermal decomposition of the fuels in automobiles. Benzene, trichloroethylene, and tetrachloroethylene et al. are well known as their own toxicities in the atmospheric environment. The unsaturated aliphatic hydrocarbons such as propylene, 1-butenes, and isoprene et al. are easily reacted with nitrogen oxides (NOx) under sunlight to produce the photochemical smog. Recently, the fine particulates of which the particle size is less than 1micrometer are formed from the VOC in the atmospheric environment. It can be assumed that the contents of fine particulates will lead to serious toxic effect to the human health.

Motor vehicles are one of main sources of air pollution in urban area, and the control of vehicle emission source has been made effort for many decades (Kirchstetter et al., 1999). The reduction of ambient concentrations of pollutants from automobiles has been accomplished to a certain extent by many effective methods (Lawryk et al., 1996), and the search for cleaner alternative fuel was a problem that has been paid much attention in many countries around the world (Gaffney et al., 1990). Oxygenated blends have been proposed as a solution for reducing of hydrocarbons, carbon monoxide (CO), and nitrogen oxides (NOx) in the emission of vehicle exhaust gas (Chang et al., 1991, Gaffney et al., 1997).
Moreover, entering the 21st century, the natural resource of petroleum is being exhausted and that leads to the use of renewable fuel as an alternative one for future. Some new pollution is expected to produce from this source, especially from alcohol fuel, one of most popular oxygenated fuel, which has been used popularly in Brazil, and been paid much attention in many countries around the world.

In the previous publication (Imamura et al., 1999), a survey on the ambient levels of 48 saturated and unsaturated, 10 aromatic, and 15 halogenated hydrocarbons, and 4 oxygenated compounds was conducted in Brazil, Los Angeles (the United States of America), and Osaka (Japan). The contribution ratios of hydrocarbon components to the urban pollution level were estimated. The methyl-tert-butylether has been used as an additive of gasoline in USA, and ethanol in Brazil, respectively.

The Republic of Vietnam is one of the countries making especially remarkable industrialization in the Southeast Asian. The numbers of motorcycles are increasing rapidly in the cities, and as a result, it is apprehensive for the influence of exhaust gas from motorcycles to the human health. The investigation on ambient air pollution in Hanoi City and Ho Chi Minh City has been carried out for the past 6 years (2000-2005 years). In this report, we described the results of concentrations of volatile organic compounds (VOC) such as 40 saturated and unsaturated aliphatic hydrocarbons, 10 aromatic hydrocarbons, and 16 halogenated hydrocarbons in Hanoi and Ho Chi Minh City during the six years.

MATERIALS AND METHODS

1. Ambient air sampling

A stainless steel canister (Entech Instruments Inc., 1000ML FSL Canister) was used for sampling. The canister cleaned and then evacuated to less than 0.1kPa (absolute pressure) was connected with the flow controller (Entech Instruments Inc., P/N 01-39-RS-2) for collecting sample at flow rate of 80ml/min in 10 min.

2. Sampling site and time

Sampling sites are listed in Table 1.

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Site description in Hanoi, Aug. 5 in 2005</th>
<th>Site description in HCM City, Aug. 11 in 2003</th>
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<tbody>
<tr>
<td>1</td>
<td>Cau Moi Bridge</td>
<td>Nguyen Thi Minh Khai and Ly Thai To</td>
</tr>
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<td>2</td>
<td>De La Thanh and Nguyen Chi Thanh</td>
<td>Nguyen Thi Tan, near bridge</td>
</tr>
<tr>
<td>3</td>
<td>Lieu Gai Street, near intersection of Hoang Hoa Tham and Lieu Gai</td>
<td>Hong Bang, near monitoring station</td>
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<tr>
<td>4</td>
<td>Ton Duc Thang and Kham Thien</td>
<td>Cach Mang Thang 8 and Ly Thuong Kiet</td>
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<td>Truong Quoc Dung, in the garden of ITTEP</td>
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<td>6</td>
<td>Tran Nhan Tong and Ba Trieu</td>
<td>Nga san Go v ap</td>
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<td>7</td>
<td>Tran Hung Dao and Du Luan</td>
<td>Cu danh Dan Quan, near monitoring station</td>
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<tr>
<td>8</td>
<td>Le Hong Phong and Dien Bien Phu</td>
<td>Nga Tu Hang Xanh</td>
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<td>Dien Bien intersection</td>
<td>Dien Bien Phu, near DOSTE</td>
</tr>
<tr>
<td>10</td>
<td>Hanoi Opera House</td>
<td>The meteorological station in Vung Tau City</td>
</tr>
</tbody>
</table>

3. Preparation of the samples for analysis after sampling:

After field sampling, the inner pressure of canister was checked and humid zero gas (nitrogen) was introduced into the canister up to 30.0psi pressure (Ashcroft Corp.: precision of barometer is 0.25%). The concentration of ambient air sample would be calculated from the dilution factor (D) deducted from (P_e/P_i) ratio where the pressures before and after introducing nitrogen were (P_i) and (P_e) respectively.

4. Preparation of the standard

4.1 Gasoline standard gas
Gasoline standard gas was prepared in 2L-round bottom flask (Supelco Co.: dilution bottle 2.0L) equipped with Mininert Valve (Supelco Co.). An aliquot of 20 μl liquid gasoline was injected through the valve to be vaporized in the flask. The flask was heated at 60°C for an hour following the storage at room temperature. After 150μg of the pure water (Wako Pure Chemical Industries) was injected to the cleaned and evacuated canister (Restec Corp.: SilicoCan Canister 6L volume), aliquot of 5ml standard gas prepared in the flask was introduced to the canister by gas tight syringe (Hamilton Corp. : 10ml volume). The canister was pressurized by humid zero gas (nitrogen) to be 14.7psi (relative pressure) of diluted standard gas. Four types of gasoline (Gasoline 83, 90, 92, 95) purchased from Ho Chi Minh City (Vietnam) were used for gasoline standard gas.

4.2 Gaseous standard

TO-14 39-components standard gas (Scotty Specialty Gases: 100ppbv each) was used for preparing standard gas in 6L stainless steel canister (0.05-5.0ppbv concentration for each components, 30psi) using by pressure dilution method for calibration. VOC 55-components standard gas (Spectra Gas: 1 ppm each) was also used for preparing standard gas in 6L stainless steel canister (0.1-22.4ppbv concentration for each components, 30psi).

5. Analytical method

A certain volume of standard gas and/or sample gas in a canister was loaded on the pre-concentrator (Entech Model 7000) before analysis by GC/MS. The pre-concentration procedures include 3 modules (glass beads, Tenax TA, fused silica capillary column (0.53mm i.d.), and sample injection was heated up to 60°C.

The gas chromatograph (HP6890 series) was used with HP-1 (1%cross-linked methyl silicone) column (60m long x 0.32mm diameter, 1.0μm in film thickness). Carrier gas was He 1.1ml/min (5.5psi at 40°C). Column temperature was held at 40°C for 4 min, programmed to 150°C at 5°C/min, and then to 220°C at 20°C/min, and finally hold at 220°C C for 2 min.

The mass spectrometer was a HP 5973 MSD and data was acquired by the scan mode. The components were determined on the basis of mass chromatography of their diagnostic ion species. An aliquot of 400ml volume of prepared standard gases (39 components of TO-14 and 55 hydrocarbons) was analyzed to make calibration curve.

6. Contribution of gasoline components to ambient air

The contribution (C(%)) of components of gasoline to ambient air were estimated on the basis of the concept of “dilution factor (df)” applied in the case of mixture of mass spectra (McLafferty et. al., 1974 and Pesyna et. al., 1976), where a component of hydrocarbons and it’s concentration was corresponding to a mass number and it’s intensity, respectively. The hydrocarbon pattern of a sampling site or gasoline, the relationship between the components and their concentrations was calculated as the highest concentration of the component is normalized as 1,000. The contribution of gasoline to ambient air is obtained by comparison with hydrocarbon pattern of gasoline (g) and that of ambient air (a). The concentration ratio (pi) between the same component (ai) of sample air and that (gi) of gasoline was calculated as following equation :

\[ \rho_i = \frac{C_{ai}}{C_{gi}} \]

where Cai and Cbi were the normalized concentrations of ai and gi, respectively. The pi was arranged in ascending order, and the df was the average of minimum three values of pi. The C(%) was estimated as follows:

\[ C(\%) = df \times \frac{100 \times \sum C_{gi}}{\sum C_{aj}} \]

The ambient air levels of the components higher than 0.1ppbv were taken in consideration, because the determination limits of this analytical method for all most of components were less than 0.05ppbv.

RESULTS AND DISCUSSION

Comparison of pollution levels at different sampling sites

Relative standard deviations (RSDs) of concentration levels of each component among the sampling sites were estimated. Except for some of halogenated hydrocarbons, the concentration
Fig. 1-1 The ambient levels of aliphatic and aromatic hydrocarbons in Hanoi and Ho Chi Minh City

1: Propylene
2: n-Propane
3: Isobutane
4: 1-Butene
5: 1,3-Butadiene
6: n-Butane
7: trans-2-Butene
8: cis-2-Butene
9: 3-Methyl-1-butene
10: Isopentane
11: 1-Pentene
12: n-Pentane
13: Isoprene
14: trans-2-Pentene
15: cis-2-Pentene
16: 2-Methyl-2-butene
17: 2,2-Dimethylbutane
18: Cyclopentene
19: Cyclopentane
20: 4-Methyl-1-pentene
21: 2,3-Dimethylbutane
22: 2-Methylpentane
23: 3-Methylpentane
24: n-Hexene
25: trans-2-Hexene
26: cis-2-Hexene
27: Methylcyclopentane
28: Cyclohexane
29: 2,4-Dimethylpentane
30: 2-Methylhexane
31: 2,3-Dimethylpentane
32: 3-Methylhexane
33: 3,4-Dimethylpentane
34: 2,3,4-Trimethylpentane
35: 2-Methylheptane
36: 2-Methylpentane
37: 2-Methylheptane
38: 3-Methylpentane
39: n-Octane
40: n-Nonane
41: Benzene
42: Toluene
43: Ethylbenzene
44: m,p-Xylene
45: Styrene
46: o-Xylene
47: Isopropylbenzene
48: Propylbenzene
49: 1,3,5-trimethylbenzene
50: 1,2,4-Trimethylbenzene

variations of the compounds were rather large depending on the sampling sites. The RSDs of those compounds were more than 50%. These results seem to be caused by the inherent difference in the emission sources, which strongly reflect to located-pollution levels.

The ambient levels of saturated and unsaturated aliphatic (40 components) were shown in Fig.1-1 by their number of 1 to 40. Among saturated aliphatic compounds, n-butane, isopentane, n-pentane, and 2-methylpentane were the abundant compounds at all sampling sites. The concentrations of isopentane, the most abundant one were in the range of 146-0.88ppbv, and the average value was 48ppbv in Hanoi, 34ppbv in Ho Chi Minh City, respectively. The levels of n-butane were in the range of 29-0.75ppbv, and the average value was 7.6ppbv in Hanoi, 8.2ppbv in Ho Chi Minh City, respectively. The levels of n-pentane were in the range of 40-0.63ppbv, and the average value was 13ppbv in Hanoi, 10ppbv in Ho Chi Minh City, respectively. The levels of 2-methylpentane were in the range of 47-0.18ppbv, and the average value was 16ppbv in Hanoi, 11ppbv in Ho Chi Minh City, respectively.

Among unsaturated aliphatic compounds, propylene, 1-butene, trans- and cis-2-butene, 2-methyl-2-butene, and 1,3-butadiene were the abundant compounds at all sampling sites. The levels of
Fig. 1-2 The ambient levels of halogenated hydrocarbons in Hanoi and Ho Chi Minh City

The figure of the right side is Hanoi and that of the left side is Ho Chi Minh City

1: Freon-12 2: Freon-11 3:Freon-113 4: Freon-114 5: Chloromethane
6: Dichloromethane 7: Chloroform 8: Carbon tetrachloride 9: Bromomethane 10: Ethylchloride
11: 1,2-Dichloroethane 12:1,1,1-Trichloroethane 13:Trichloroethylene 14: Tetrachloroethylene 15: Chlorobenzene
16: p-Dichlorobenzene

propene, the most abundant one were in the range of 62-0.13ppbv, and the average value was 16ppbv in Hanoi, 14ppbv in Ho Chi Minh City, respectively. The levels of 1-butene were in the range of 42-0.04ppbv, and the average value was 12ppbv in Hanoi, 10ppbv in Ho Chi Minh City, respectively. The levels of trans- and cis-2-butene were in the range of 23-0.06ppbv, and 21-0.05ppbv, respectively, and the average value was 8.2 and 7.8ppbv in Hanoi, 6.2 and 5.7ppbv in Ho Chi Minh City, respectively. The levels of 2-methyl-2-butene were in the range of 58-0.05ppbv, and the average value was 19ppbv in Hanoi, 13ppbv in Ho Chi Minh City, respectively. The levels of 1,3-butadiene were in the range of 18-0.03ppbv, and the average value was 5.1ppbv in Hanoi, 4.1ppbv in Ho Chi Minh City, respectively. The levels of these unsaturated aliphatic hydrocarbons were relatively high at the roadside. It can be assumed that these components were affected from exhaust gas of automobiles.

The ambient levels of aromatic hydrocarbons (10 components) were shown in Fig. 1-1 by their number of 41 to 50. Among aromatic compounds, benzene, toluene, m,p-xylene, o-xylene, and 1,2,4-trimethylbenzene were the abundant compounds at all sampling sites. The levels of toluene, the most abundant one, were in the range of 169-1.0ppbv, and the average value was 27ppbv in Hanoi, 39ppbv in Ho Chi Minh City, respectively. The average values of benzene, m,p-xylene, o-xylene, 1,2,4-trimethylbenzene were 14, 22, 8.3, 13ppbv in Hanoi, and 12, 21, 8.2, 8.8ppbv, respectively.

The ambient levels of halogenated hydrocarbons (16 components) were shown Fig. 1-2. Among halogenated compounds, chloromethane and dichloromethane were the abundant compounds and their average values were 0.75 and 0.43ppbv in Hanoi, and 4.0 and 0.83ppbv in Ho Chi Minh City, respectively. The solvent of dichloromethane had been widely utilized in Vietnam, because the levels of the other solvents of 1,1,1-trichloroethane, trichloroethylene, and tetrachloroethylene were less than 0.05ppbv in the urban atmosphere. Some of halogenated hydrocarbons showed the almost same levels in all sampling sites: the average of freon 11 was 0.23ppbv (RSD 1.6%) in Hanoi, and 0.23ppbv (RSD 13%) in Ho Chi Minh City, that of freon 12 0.56ppbv (RSD 26%) in Hanoi, and 0.54ppbv (RSD 31%) in Ho Chi Minh City, that of carbon tetrachloride 0.11ppbv(RSD 6.2%) in Hanoi, and 0.09ppbv (RSD 18%) in Ho Chi Minh City, and that of freon 113 0.08ppbv (RSD 11%) in Hanoi, and 0.07ppbv (RSD 8.2%) in Ho Chi Minh City. According to the Montreal Protocol, these four halogenated compounds were forbidden in manufacture as well as utilization from the
end of 1995. After mid-1998, the levels of these compounds were remained almost uniform (Imamura et al., 1995; Hasegawa et al., 2000) in entire global atmosphere.

**Gasoline fuel contents**

Contents of commercially available gasoline in Vietnam were examined. Four types of commercial gasoline were Gasoline 83, 90, 92 and 95 of which the number is referred to the octane value. Each concentration of components contained in the gasoline was normalized, as that of isopentane and/or toluene was 1000, and the relative intensities of each component were shown in Fig. 2. The results show that isopentane and/or toluene makes up the main content and their contents were the same level in four types of gasoline. Except for two components, saturated aliphatic hydrocarbons of n-butane, n-pentane, 2-, and 3-methylpentane, 2-methylpentane, and n-hexane, unsaturated aliphatic hydrocarbons of trans-2-penene and 2-methyl-2-butene, and aromatic hydrocarbons of benzene, toluene, m,p-xylene, o-xylene, and 1,2,4-trimethylbenzene were the abundant components in gasoline. The patterns of relative intensities of hydrocarbon components in the four types of gasoline were in good agreement with those in the ambient atmosphere as described previously. It was suggested that the evapoemission (emission of evaporated gasoline from a automobile fuel tank) was one of the reasons why aromatic and saturated aliphatic hydrocarbons showed the highest content in the urban atmosphere of the cities.

**Estimating the contribution of gasoline to the atmospheric pollution**

In the atmosphere, pollution consists of many pollutants emitted from vehicle exhaust, that is, mobile source, and from industrial manufacture, that is, stationary source, and the natural source such as terpene. It is important to estimate the contribution of these sources to take administrative countermeasures to reduce the specific pollutants in the atmosphere. Recently, it has been reported that concerning to the pollution of hydrocarbons in the atmosphere, not only the automobile exhaust (mobile source) contributes to atmospheric pollution, but emission from fuel tank (evapoemission) also plays an important role. The other emission sources of hydrocarbons taken into consideration in this report were the liquefied petroleum (LP) and the components (Exhaust) of hydrocarbons exhausted from the automobiles.

The contribution of the hydrocarbon components from emission sources to the atmospheric pollution was estimated for each sampling site by using results of gasoline components, which were discussed in previous section. The results were shown in Table 2. The total hydrocarbons, the sum
of the values of hydrocarbon components at the sampling site were also cited. The contribution ratios of Gasoline 83 were less than 30% at all sampling sites. This means that Gasoline 90, 92, and 95, the type of the high combustion efficiency were popular in the market of Vietnam. The contribution ratio of those three types of gasoline were estimated to be more than 30% at the sampling sites of the roadside, of which the total hydrocarbons were more than 200 ppbv. On the contrary, the contribution ratio of LP&Exhaust increased up to more than 35% at the sampling sites of which the total hydrocarbons were less than 100 ppbv. These results indicated that the contribution of evapoemission of gasoline that means to say, the unburned fuel was estimated to be up to the half of value of the hydrocarbons in the urban atmosphere.

Table 2-1  Contribution of Gasoline Components to Ambient Air Hydrocarbons in Hanoi

<table>
<thead>
<tr>
<th>Sampling Sites</th>
<th>Total Hydrocarbons (ppb)</th>
<th>Gasoline 95 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
<th>Gasoline 92 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
<th>Gasoline 90 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
<th>Gasoline 83 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
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LP : Liquefied petroleum of which the main components are propane, n-butane and isobutane
Exhaust : the components of hydrocarbons exhausted from the automobiles, and contained the unsaturated hydrocarbons of propylene, 1-butene, 1,3-butadiene, and so on.

Table 2-2  Contribution of Gasoline Components to Ambient Air Hydrocarbons in Ho Chi Minh City

<table>
<thead>
<tr>
<th>Sampling Sites</th>
<th>Total Hydrocarbons (ppb)</th>
<th>Gasoline 95 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
<th>Gasoline 92 Contribution (%)</th>
<th>LP&amp;Exhaust Contribution (%)</th>
<th>Gasoline 90 Contribution (%)</th>
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CONCLUSIONS

The ambient levels of 40 saturated and unsaturated aliphatic hydrocarbons, 10 aromatic hydrocarbons, and 16 halogenated hydrocarbons were determined in Hanoi, and Ho Chi Minh City. The results obtained were as follows:

1. Relative standard deviations of ambient levels for all aliphatic and aromatic hydrocarbons were more than 80%, because of the contribution of their different sampling site from the emission source.
2. An ambient level of aliphatic hydrocarbons of isopentane and aromatic hydrocarbons of toluene, and \( m,p \)-xylene were the most abundant components in both cities.

3. The levels of halogenated hydrocarbons (trichlorofluoromethane (freon 11), dichlorodifluoromethane (freon 12), 1,1,2-trichloro-1,2,2-trifluoroethane (freon 113), and carbon tetrachloride) were low and almost uniformed at all sampling sites.

4. The average values of chloromethane and dichloromethane utilized as solvents were 0.75 and 0.43 ppbv in Hanoi, and 4.0 and 0.83 ppbv in Ho Chi Minh City, respectively.

5. Constituents of four types of gasoline available in the market were determined for comparison, and contribution ratios of gasoline to atmospheric pollution levels were estimated to be 3-57% and 8-56%, and those of LP&Exhaust to be 8-40% and 10-32% for Hanoi and Ho Chi Minh City, respectively.

REFERENCES


