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Citation	Annual Report of FY 2006, The Core University Program between Japan Society for the Promotion of Science (JSPS) and Vietnamese Academy of Science and Technology (VAST). 2007, p. 47-54
Version Type	VoR
URL	https://hdl.handle.net/11094/13032
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INVESTIGATION ON AIR POLLUTION IN VIETNAM -VOLATILE ORGANIC COMPOUNDS IN HANOI AND HO CHI MINH-

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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 Mihn 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 1 Sampling sites and date in Hanoi and Ho Chi Minh City

Site No.	Site description in Hanoi, Aug. 5 in 2005		Site description in HCM City, Aug. 11 in 2003	
1	Cau Moi Bridge	intersection	Nguyen Thi Minh Khai and Ly Thai To	intersection
2	De La Thanh and Nguyen Chi Thanh	intersection	Nguyen Thi Tan, naer bridge	intersection
3	Lieu Gai Street, near intersection of Hoang Hoa Tham and Lieu Giai	roadside	Hong Bang, near monitoring station	road side
4	Ton Duc Thang and Kham Thien	intersection	Cach Mang Thang 8 and Ly Thuong Kiet	intersection
5	Dai Co Viet and Giai Phong	intersection	Truong Quoc Dung, in the garden of ITTEP	ambient
6	Tran Nhan Tong and Ba Trieu	intersection	Nga sau Go vap	intersection
7	Tran Hung Dao and Le Duan	intersection	Uy Ban Nhan Dan Quan, near monitoring station	ambient
8	Le Hong Phong and Dien Bien Phu	intersection	Nga Tu Hang Xanh	intersection
9	Long Bien intersection	intersection	Dien Bien Phu, near DOSTE	road side
10	Hanoi Opera House	roadside	The meteorogical station in Vung Tau City	ambient

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) deduced from (Pe/Pi) ratio where the pressures before and after introducing nitrogen were (Pi) and (Pe) 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: 1ppm 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 (ρi) between the same component (ai) of sample air and that (gi) of gasoline was calculated as following equation :

$$\rho i = Cai / Cgi$$

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

$$C(\%) = df \times 100 \times \Sigma Cgi / \Sigma Caj$$

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

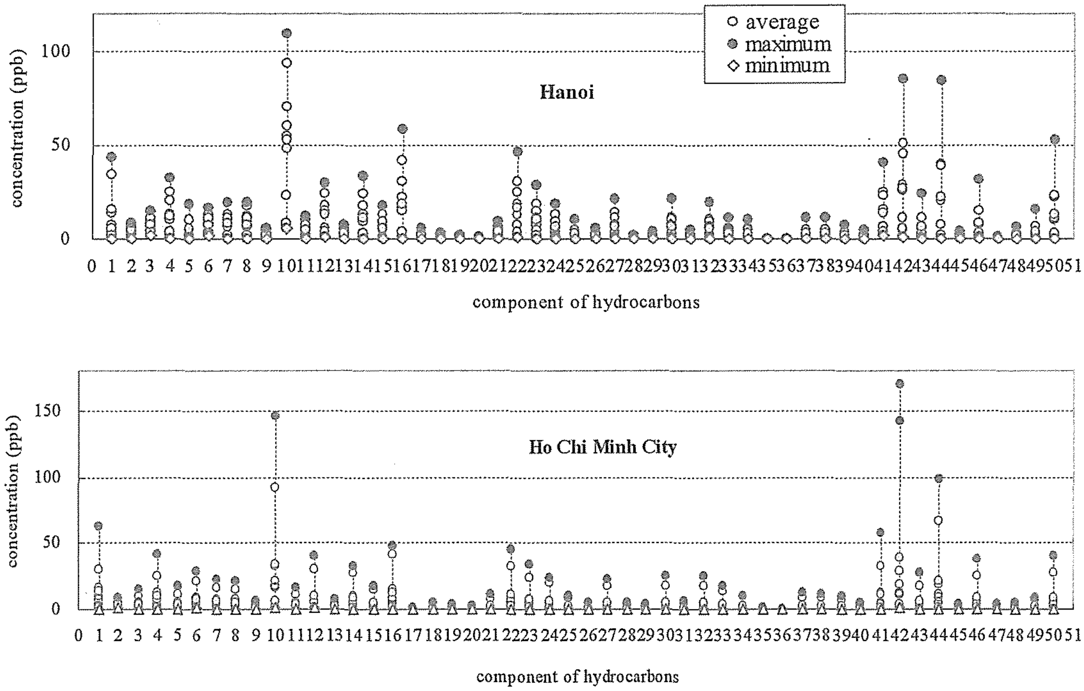

Fig. 1-1 The ambient levels of aliphatic and aromatic hydrocarbons in Hanoi and Ho Chi Minh City

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-Methy-1-pentene 21: 2,3-Dimethylbutane 22: 2-Methylpentane 23: 3-Methylpentane 24: *n*-Hexane
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 34: Methylcyclohexane
35: 2,2,4-Trimethylpentane 36: 2,3,4-Trimethylpentane 37: 2-Methylheptane 38: 3-Methylheptane
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 seam 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-methylentane 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, proylene, 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

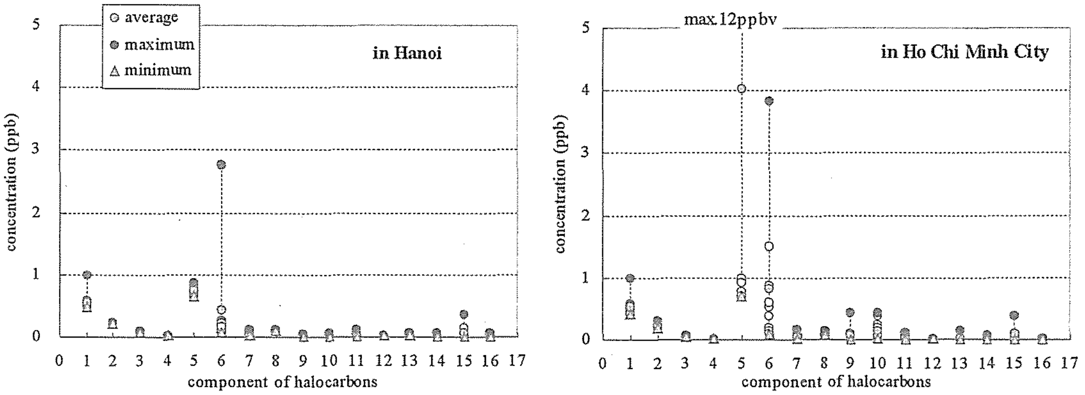

Fig. 1-2 The ambient levels of halogenated hydrocarbons in Hanoi and Ho Chi Minh City
The figure of the right side is Hanoi and that of the left side is Ho Chi Mihn City
1: Freon-12 2: Freon-11 3 :Freon-113 4: Freon-114 5: Chloromethane
6: Dichloromethane 7: Chloroform 8: Carbon tetrachloride 9: Bromomethane 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 therange 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 (10components) 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 (16components) 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 Mihn City, that of freon 12 0.56ppbv (RSD 26%) in Hanoi, and 0.54ppbv (RSD 31%) in Ho Chi Mihn City, that of carbon tetrachloride 0.11ppbv(RSD 6.2%) in Hanoi, and 0.09ppbv (RSD 18%) in Ho Chi Mihn City, and that of freon 113 0.08ppbv (RSD 11%) in Hanoi, and 0.07ppbv (RSD 8.2%) in Ho Chi Mihn City. According to the Montreal Protocol, these four halogenated compounds were forbidden in manufacture as well as utilization from the

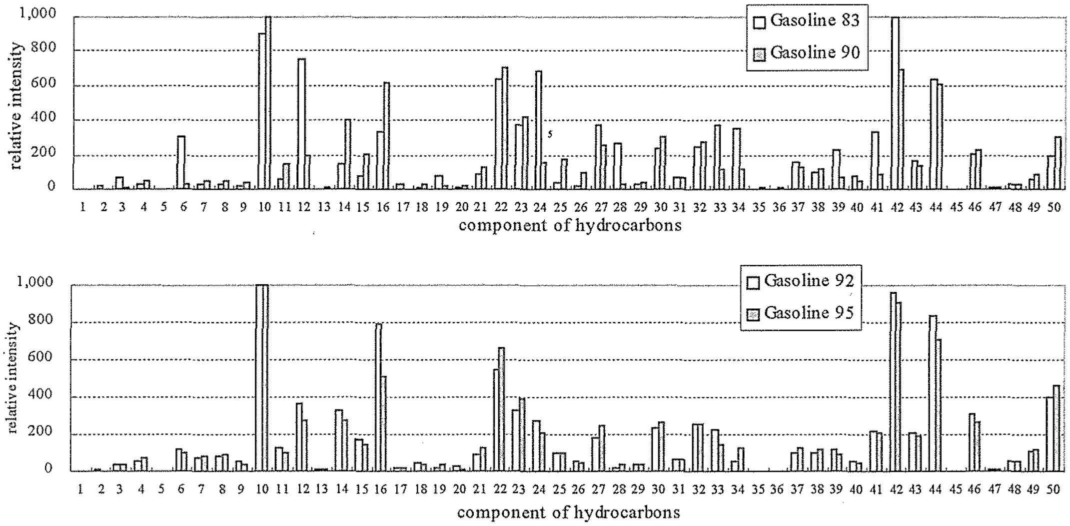

Fig. 2 Relative intensities of component of hydrocarbons in various gasoline available in Vietnam
Numbers of component of hydrocarbons were cited in Fig.1

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 200ppbv. 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 100ppbv. 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.

Table2-1 Contribution of Gasoline Conponents to Ambient Air Hydrocarbons in Hanoi

Sampling Sites	Total Hydrocarbons (ppb)	Gasoline 95		Gasoline 92		Gasoline 90		Gasoline 83	
		Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)
1	1,032	57	10	48	11	44	11	27	10
2	368	41	18	37	18	31	18	19	18
3	59	20	35	20	35	22	36	10	35
4	522	53	7.5	46	7.7	37	8.3	24	7.7
5	387	46	22	39	22	25	23	23	22
6	655	54	11	40	11	36	12	24	11
7	255	19	15	18	15	20	15	11	15
8	124	32	13	32	13	18	14	19	13
9	62	3.7	39	2.7	39	3.1	39	7.3	39
10	21	7.9	40	4.8	40	8.1	40	3.5	40

Table2-2 Contribution of Gasoline Conponents to Ambient Air Hydrocarbons in Ho Chi Minh City

Sampling Sites	Total Hydrocarbons (ppb)	Gasoline 95		Gasoline 92		Gasoline 90		Gasoline 83	
		Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)	Contribution (%)	LP&Exhuast (%)
1	1,262	50	13	51	13	34	14	18	13
2	942	50	10	56	10	40	10	23	10
3	131	30	22	35	22	25	22	22	22
4	311	46	16	51	16	40	17	16	16
5	56	18	26	22	26	23	26	9.4	26
6	180	49	14	49	14	41	14	14	14
7	37	19	30	25	30	22	30	13	30
8	152	27	18	23	18	22	18	9.3	18
9	39	25	18	28	18	21	18	16	18
10	10	11	32	10	32	7.8	32	14	31

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.

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 0.43ppbv in Hanoi, and 4.0 and 0.83ppbv 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.

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