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SHORT COMMUNICATION

Determination of Non-Methane Hydrocarbon Emission Factors from Vehicles in a Tunnel in Seoul in May 2000

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Abstract−Measurements of non-methane hydrocarbons (NMHC) were performed at the entrance and exit of the Sangdo tunnel to estimate emission factors (EF) of NMHC from vehicles in May 2000. About 50 species were analyzed by a combined GC/FID and GC/MS system. Ethylene was the most abundant compound, followed by n-butane and acetylene, respectively. Based on the measurement data, the real world vehicular EF in Seoul was estimated. The highest EF value was 89.8 mg (veh-mile)$^{-1}$ for n-butane, followed by ethylene and toluene.

Key words: Non-Methane Hydrocarbons, Vehicular Emission Factors, Tunnel Measurement, Seoul

INTRODUCTION

Road traffic is one of the most significant sources of primary air pollutants in urban areas. Hydrocarbons from vehicular emissions have been recognized as one of the major contributors to the formation of photochemical smog and the detrimental effects on human health. Therefore, it is very important to study vehicular emissions in order to understand atmospheric hydrocarbon concentrations and levels.

Vehicular emissions of primary air pollutants can be described by the emission factor (EF), defined as the emitted mass of a chemical compound per distance per vehicle. EF of vehicles depends on many different factors such as design of the engine (gasoline with or without catalytic converter, or diesel), driving conditions (acceleration, etc), and fuel used. The EF can be determined by exhaust measurements from single vehicles in a dynamometric test. However, the EF of a large number of vehicles has to be measured to obtain representative results for actual road traffic emissions.

Road traffic emissions can also be determined by exhaust measurements of driving vehicles [Bailey et al., 1990] or in tunnel measurements. The compositions of the hydrocarbon species in the tunnel air are broadly representative of a large number of vehicles and fuel types used in urban areas. Additionally, tunnel measurements have the advantage of quickly obtaining composite samples [Nelson and Quigley, 1984].

We made a tunnel measurement in February 2000 at Sangdo tunnel located in the southern part of Seoul [Na and Kim, 2000]. Since this tunnel is located in Seoul, the capital of Korea, this measurement was expected to help further the understanding of the characteristics of vehicle emissions in urban areas of Korea. However, during that measurement, only one sampling point was set in the tunnel; thus, accurate estimation of emission factors was hard to do from the data. The present study was conducted in May 2000 and two sampling points were set up inside the tunnel to make mass balance calculation possible.

The main purpose of this study is the estimation of the EF of a large number of individual volatile organic compounds in Seoul. The EF was calculated for the total vehicle classes without any classification of vehicles. Here we described the results of the EF calculated from 10 samples. We further compared the result with other results from the previous tunnel studies conducted abroad.

EXPERIMENT

Heavy traffic usually begins at 07 : 30 am and continues to 08 : 30 am. From 07 : 00-07 : 30 am, the traffic was light. The composition and number of vehicles passing through the tunnel were determined by direct counting. A total of 6,076 vehicles were counted during the study period. The traffic counts were split into four classes: 62.8% were identified as gasoline-fueled vehicles, 24.0% as diesel-fueled vehicles, 11.6% as butane-fueled vehicles, and 1.7% gasoline-fueled motorcycles. This composition is in good agreement with the data of vehicle registration in Seoul [Na and Kim, 2000]. The range of fluctuation in vehicle compositions was within 10% in the tunnel during the experiments.

Measurement was performed from 29 to 31 May 2000 in the slightly downhill grade tunnel which carries traffic in one direction on two lanes. Detailed information on the tunnel configuration is given at Na and Kim [2000]. Grab sampling was conducted by using 6 L SUMMA polished stainless steel canisters under high vacuum of 10^{-4} Torr. Sampling runs of 30 min duration were conducted simultaneously between 07 : 00 am and 08 : 30 am at the entry and the exit sites. During this time period, passing vehicles experienced various driving conditions with starting, moving, and stopping. The entry site is 50 m from the tunnel entrance. The sampling sites were separated by 250 m. The tunnel is not equipped with any forced ventilation system. A total of ten samples were collected, five samples per each sampling point. The range of sampling time was chosen to represent mixed characteristics of various driving conditions. Table

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Table 1. Individual runs in the Sangdo tunnel study

Run	Time	Date	Number of vehicles	Temperature (°C)	Humidity (%)
1	07 : 00-07 : 30	5/29/2000	1,220	17.8	54
2	07 : 30-08 : 00	5/29/2000	1,095	17.7	53
3	07 : 00-07 : 30	5/30/2000	1,410	15.5	60
4	07 : 30-08 : 00	5/31/2000	1,023	16.8	56
5	08 : 00-08 : 30	5/31/2000	1,302	17.5	54

1 gives the conditions for each tunnel run in the study. Sampling position is 1.8 and 2.0 m from the ground level and passing vehicles, respectively. Air speed in the tunnel was monitored by a portable anemometer at the sampling site 300 m away from the entrance to the tunnel.

The canisters were analyzed by a GC/FID and GC/MS system at Korea Institute of Science and Technology (KIST). The GC/FID was used to quantify C_2-C_3 hydrocarbons, and the GC/MS was used to identify C_2-C_9 hydrocarbons and quantify C_4-C_9 hydrocarbons. For the analysis of C_2 and C_3 hydrocarbons, a GC/FID (STAR 3600CX, Varian, USA) was used. Separation was achieved by a 60 m long, 0.32 mm I.D., 3.0 μm film thickness RTX-1 column. Initial oven temperature was −60 °C for 7 min and raised to 70 °C at the rate of 10 °C min^{-1} and then the oven was heated to the final temperature, 220 °C, at the rate of 7 °C min^{-1} and was kept at 220 °C for 15 min. Flow rate of the carrier gas, N_2 (99.9999%) was 3 ml min^{-1} and that of the detector make up gas (N_2) was 30 ml min^{-1}. Flow rates of H_2 and air were 19 and 250 ml min^{-1}, respectively. Detector was heated to 250 °C. Sample was concentrated for 5 min at a flow rate of 40 ml min^{-1}. Liquid nitrogen was used as the coolant for the cryogenic trap, and temperature of which was −150 °C for 5 min. Then samples were desorbed at 200 °C for 2.5 min.

C_4-C_9 hydrocarbons were identified by means of a combined gas chromatography/mass spectrometry (GC/MS) system (3400CX GC & Saturn 2000 MS, Varian, USA). A sample preconcentration trap for the standards and samples was packed with 60-80 mesh glass beads. To concentrate the hydrocarbons, an aliquot of a canister air was transferred into a trap immersed in liquid nitrogen at the flow rate of 20 ml min^{-1} for 5 min. The total volume concentrated was 100 ml. The concentrated sample was revolatized by heating the trap up to 170 °C and then held for 11.3 min. Hydrocarbons were separated in a 60 m long, 0.32 mm I.D., 1.0 μm film thickness DB-1 column (J&W Scientific, USA), and subsequently analyzed by the GC/MS. The GC oven program used was as follows: initial temperature 0 °C for 7 min, rising at 15 °C min^{-1} up to 170 °C where it remains for 6 min. Calibration standards (Scott, USA) were used to establish the retention times and detector responses of the GC/MS.

Precision, as determined from five replicate analyses of the standards and samples, is within 15% for the compounds at the concentrations above 5 ppbC and the lower quantifiable limits were between 0.1 and 0.5 ppbC depending on component for the 100 ml of sample concentrated. The percent difference of the concentrations of VOC for the six duplicate samples measured during the period of this study is less than 35%. The accuracy was demonstrated through a comparison analysis between KIST and Atm AA, an environmental consulting laboratory, USA on four of the same samples. The results show that the relative errors calculated on the basis of Atm AA range from 3 to 49%. Details are given at Na and Kim [2001].

RESULTS AND DISCUSSION

1. Determination of Emission Rate

Emission factors for traffic within a tunnel can be calculated per unit of fuel consumed or per vehicle-distance-traveled [Fraser et al., 1998]. In the present study, we adopted EF with the emitted mass of a compound per vehicle-mile since the estimated EF values will be used in a model using the unit mile. The method of extracting emission rates from tunnel measurements has been described by Pierson et al. [1990]. Briefly, one samples simultaneously at two points, one near to the exit and the other at the tunnel entrance. To determine the mass of a constituent i produced by passing vehicles in the tunnel, we assumed that an element volume of the tunnel is a steady-state plug flow reactor. The crucial feature of this reactor is that the composition of the fluid varies from point to point along a flow. Thus, the material balance in the tunnel with the generated mass M_i (in mg), the traffic count N (number of vehicles), and the tunnel length L (distance between sampling sites, 250 m in this study) for the emission rate of E_i in mg (veh-mile)$^{-1}$ for a species i of interest is given:

$$E_i = M_i/(NL). \quad (1)$$

2. General Characteristics

The average total NMHC level at the exit of the tunnel (2,559 ppbC) was twice as high as that at the entrance (1,151 ppbC). At both points, alkanes are the most abundant, followed by aromatics, and alkenes. Additionally, the compositions of the hydrocarbon classes are similar to each other suggesting that the NMHC concentrations at the two points are primarily influenced by vehicle exhausts.

The average concentrations and composition with their standard deviations of NMHC at the entrance and exit of the Sangdo tunnel are given in Table 2 along with the composition data measured in February 2000 in the same tunnel [Na and Kim, 2000]. The most abundant compounds observed in the tunnel were ethylene, followed by n-butane, acetylene, and i-butane on the basis of averaged concentrations (ppb) of the two points. In weight percent basis, butane (11.7%) is the most abundant in the tunnel. The most prominent difference between the February and May measurements is the mass fractions of propane. During the wintertime (from November to March), 5 to 30 wt% of propane is added to the butane fuel which is used for the some types of vehicles such as taxicabs and recreational vehicles. However, during the remainder of the year, butane fuel contains no propane. This is the main reason for that difference. Accordingly, the seasonal pattern of fuel seems to affect

Table 2. Concentrations and compositions measured in the Sangdo tunnel in May 2000

	This study (May 2000)				Composition in Feb. (wt%)
	Concentrations (ppb)		Compositions (wt%)		
	Entrance of tunnel	Exit of tunnel	Entrance of tunnel	Exit of tunnel	
Ethane	7.5±3.9	12.6±5.7	1.6±0.4	1.4±0.5	3.4±1.2
Propane	8.6±4.3	11.8±5.3	2.8±1.1	2.3±1.3	6.6±1.5
n-Butane	31.8±26.8	75.8±69.4	11.0±1.0	12.4±1.2	12.7±2.1
i-Butane	17.0±15.0	36.7±35.7	5.8±0.7	5.9±0.8	7.2±0.9
n-Pentane	6.3±5.2	14.5±13.7	2.7±0.5	2.8±0.4	2.8±0.3
i-Pentane	7.9±6.7	17.9±17.7	3.4±0.6	3.3±1.0	3.1±0.4
2-Methelpentane	4.9±3.5	11.7±11.4	2.7±0.2	2.7±0.2	2.6±0.2
3-Methylpentane	1.0±1.0	8.8±12.9	0.5±0.3	1.5±1.2	0.6±0.1
2,2-Dimethylbutane	0.7±0.7	1.6±2.1	0.3±0.2	0.3±0.2	0.1±0.1
2,3-Dimethylbutane	4.0±3.2	9.8±10.3	2.1±0.3	2.2±0.3	1.8±0.3
n-Hexane	4.1±2.6	8.8±8.8	2.3±0.6	2.1±0.3	1.7±0.4
2-Methylhexane	3.2±2.3	7.7±8.2	2.0±0.2	2.0±0.2	0.6±0.4
3-Methylhexane	1.5±0.8	2.6±2.8	1.2±0.8	0.7±0.1	0.1±0.1
2,3-Dimethylpentane	2.4±2.2	6.1±6.3	1.4±0.2	1.6±0.2	1.2±0.3
2,4-Dimethylpentane	0.5±0.5	1.4±1.4	0.3±0.1	0.4±0.1	0.2±0.1
n-Heptane	2.1±1.9	5.1±5.7	1.2±0.1	1.3±0.3	1.1±0.2
2,2,4-Trimethylpentane	0.5±0.6	1.5±1.6	0.2±0.2	0.4±0.1	0.2±0.1
2,3,4-Trimethylpentane	0.3±0.2	1.1±0.7	0.4±0.3	0.4±0.3	0.1±0.1
2-Methylheptane	0.7±0.7	1.5±1.6	0.4±0.2	0.5±0.0	0.3±0.2
3-Methylheptane	0.8±1.0	2.2±2.4	0.4±0.4	0.6±0.1	0.4±0.2
Octane	0.5±0.5	1.3±1.1	0.3±0.3	0.4±0.1	0.4±0.1
Nonane	0.8±0.5	1.5±1.5	0.7±0.2	0.5±0.1	0.3±0.2
Ethylene	52.2±32.2	93.6±47.5	9.6±1.1	9.3±2.9	9.4±1.4
Propylene	15.3±9.1	27.3±13.3	4.3±0.5	4.1±1.3	4.2±0.7
1-Butene	8.3±7.8	21.4±26.6	2.8±0.7	3.0±1.3	2.8±1.3
t-2-Butene	5.1±4.7	11.6±13.3	1.6±0.5	1.5±0.5	0.7±0.1
c-2-Butene	2.5±2.4	5.6±6.1	0.7±0.4	0.8±0.2	0.5±0.1
1-Pentene	1.0±1.1	2.7±3.1	0.4±0.1	0.4±0.2	0.2±0.1
Isoprene	0.8±0.8	1.5±1.7	0.3±0.2	0.2±0.1	0.1±0.1
t-2-Pentene	1.9±1.8	4.0±6.4	0.8±0.1	0.4±0.5	0.5±0.1
c-2-pentene	2.0±1.8	5.2±5.8	0.8±0.1	0.9±0.3	0.2±0.1
2-Methyl-2-butene	2.8±2.6	4.6±3.0	1.1±0.2	1.0±0.4	0.7±0.2
Acetylene	19.5±10.8	34.4±19.6	3.4±0.8	3.0±0.8	4.1±1.4
Cyclopentane	0.8±0.8	2.0±2.1	0.3±0.2	0.3±0.2	0.3±0.2
Methylcyclopentane	2.3±1.7	5.4±5.1	1.2±0.1	1.2±0.1	1.2±0.1
Cyclohexane	0.5±0.4	1.1±1.2	0.2±0.1	0.2±0.1	0.5±0.1
Methylcyclohexane	0.8±0.8	1.9±2.0	0.4±0.2	0.5±0.1	0.4±0.1
Benzene	6.3±5.4	15.0±16.0	3.0±0.4	3.0±0.4	3.4±0.2
Toluene	16.0±11.8	32.1±29.0	9.8±3.2	8.8±2.4	9.5±1.9
Ethylbenzene	1.9±1.4	4.3±4.6	1.3±0.2	1.2±0.2	1.1±0.1
m-/p-Xylene	9.1±6.0	22.1±23.9	6.4±1.0	6.3±1.0	4.5±2.4
o-Xylene	2.3±1.6	5.7±6.2	1.6±0.4	1.6±0.2	1.6±0.3
Styrene	0.5±0.8	1.2±1.4	0.3±0.4	0.3±0.2	0.0±0.0
1,2,4-Trimethylbenzene	6.7±7.4	16.8±19.0	4.5±1.5	5.0±0.9	5.3±1.8
1,3,5-Trimethylbenzene	1.7±1.4	3.4±3.7	1.3±0.4	1.1±0.2	1.1±0.3

The Feb. data are from Na and Kim [2000].

the compositions of NMHC emitted from vehicle exhausts.

3. Emission Factors from the Sangdo Tunnel

The NMHC composition in the tunnel is determined by the sources inside the tunnel and by the composition of the outside air

entering the tunnel. To examine the effects of the outside air on the inside air, we compared the abundances of major compounds of sources of non-vehicle exhaust at the exit with those at the entrance of a tunnel. In Seoul, ethane is known to be the major compound of natural gas that is usually used for heating and cooking [Na and Kim, 2000]. Since the tunnel is close to residential areas, the tunnel air can be affected by hydrocarbons emitted from the areas through air entering the tunnel. The compositions of ethane are 1.6

Table 3. Calculated emission factors of NMHC in mg/veh-mile in the Sangdo tunnel and other studies. The cells for the compounds not reported are left vacant

	Average	S. D.	Thiais*	Fort McHenry**	Tuscarora**
Ethane	6.6	4.5	15.6	8.6	5.4
Propane	8.8	6.3	4.8	1.3	1.1
n-Butane	89.8	32.3	57.2	10.0	8.2
i-Butane	39.1	17.5	24.1	1.2	1.6
n-Pentane	19.6	11.3	12.6	15.1	8.7
i-Pentane	21.9	17.4	153.0	48.7	23.6
2-Methylpentane	18.6	12.0	15.4	16.3	7.7
3-Methylpentane	19.1	22.5	9.1	9.1	4.9
2,2-Dimethylbutane	1.4	3.9		5.2	2.9
2,3-Dimethylbutane	15.4	11.5		5.9	2.3
n-Hexane	13.0	11.3	5.5	7.5	3.9
2-Methylhexane	13.0	11.3			
3-Methylhexane	−0.4	10.4		8.2	2.5
2,3-Dimethylpentane	12.2	6.4		4.9	1.9
2,4-Dimethylpentane	3.5	1.5		3.9	1.5
n-Heptane	8.9	7.7		3.9	1.6
2,2,4-Trimethylmpentane	4.7	2.0		18.0	6.2
2,3,4-Trimethylpentane	3.2	2.9		6.4	2.1
2-Methylheptane	3.3	2.0		2.3	1.3
3-Methylheptane	6.5	2.9		2.6	1.2
Octane	4.1	1.6		1.7	0.8
Nonane	2.4	3.5		1.5	0.3
Ethylene	50.1	28.1	136.5	36.8	23.9
Propylene	21.8	12.0	61.2	14.1	9.7
1-Butene	19.3	21.8			
t-2-Butene	10.4	9.8	7.7	2.0	0.8
c-2-Butene	6.3	4.7	5.7	1.5	1.3
1-Pentene	3.0	4.4	3.3	2.0	1.0
Isoprene	1.1	1.4		1.6	1.1
t-2-Pentene	1.2	8.6	6.5	3.4	1.8
c-2-Pentene	6.7	6.1	3.4	2.0	1.1
2-Methyl 2-butene	5.1	5.8	2.4	5.4	2.9
Acetylene	15.8	7.7	67.4	12.3	6.3
Cyclopentane	2.6	2.3		1.8	0.9
Methylcyclopentane	8.3	5.5			
Cyclohexane	1.5	1.4			
Methylcyclohexane	4.0	2.1			
Benzene	20.6	15.9		23.5	14.7
Toluene	50.0	28.5		44.7	22.8
Ethylbenzene	7.2	7.0		11.7	4.5
m-/p-Xylene	41.0	33.8		40.1	16.9
o-Xylene	10.8	8.6		14.6	6.5
Styrene	2.2	2.1		5.4	2.7
1,2,4-Trimethylbenzene	37.0	24.3		25.5	8.6
1,3,5-Trimethylbenzene	5.3	6.5		6.9	2.7

*: Tousty and Bonsang [2000]; **: Sagebiel et al. [1996].

and 1.4 wt%, respectively, for the entrance and exit of the tunnel. Relatively higher abundances of ethane at the entrance of the tunnel reflect the existence of the effects of non-vehicle sources of the outside of the tunnel on the tunnel air.

It has been suggested that one measure of catalytic converter efficiency is the ethylene/acetylene ratio. Since the introduction of emission controls, the emissions of ethylene and acetylene have both decreased as a fraction of total NMHC [Fujita et al., 1995]. However, the decrease is greater for acetylene because it is removed more efficiently by the catalyst than ethylene. Well maintained catalyst-equipped vehicles have ethylene/acetylene ratios of 3 or greater based on Federal Test Procedure emission tests, whereas non-catalyst cars have ratios closer to 1 [Hoekman, 1992]. For example, the ratio for Cairo, Egypt was reported to 0.75. The reason for this was explained by characteristics of vehicles without catalysts [Doskey et al., 1999]. During this study, the observed mole ratio was 2.7. This value is slightly larger than that (2.3) in February 2000 which suggests that the efficiency of catalysts is affected by ambient temperature.

The emission factor calculated from Eq. (1) is given in Table 3 along with other reported tunnel studies abroad. The EF of NMHC in this study is generally comparable to those of Thiais and Fort McHenry tunnels, but higher by about a factor of 2 in the Tuscarora tunnel [Touaty and Bonsang, 2000; Sagebiel et al., 1996].

The emission factor of n-butane is the highest value measured at 89.8 mg (veh-mile)$^{-1}$, followed by ethylene of 50.1 mg (veh-mile)$^{-1}$ and toluene of 50.0 mg (veh-mile)$^{-1}$. Note that isoprene, the main compound emitted from biogenic source, is 1.1 mg (veh-mile)$^{-1}$ of emission factor. Fort McHenry and Tuscarora tunnels also released at the same order of magnitude. It shows that isoprene is produced also by combustion of vehicle fuels.

SUMMARY

Emission factors (EF) of non-methane hydrocarbons (NMHC) are important data to estimate vehicular emissions of NMHC which contains ozone precursors and hazardous air pollutants. To develop cost-effective control strategies against NMHC levels in the air, it is essential to have accurate emission inventories of major NMHC.

Measurements of NMHC were carried out at the entrance and exit of the Sangdo tunnel to estimate emission factors (EF) of NMHC from vehicles in May 2000. About 50 species were analyzed by a combined GC/FID and GC/MS system. Ethylene was the most abundant compound, followed by n-butane, and acetylene, respectively. Based on the measurement data, the real world vehicular EF in Seoul was estimated. The highest EF value was 89.8 mg (veh-mile)$^{-1}$ for n-butane, followed by ethylene and toluene.

This result can be used as a data for source identification of the ambient NMHC in Seoul along with other emission source data such as Na et al. [2001].

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NOMENCLATURE

E_i : emission rate of chemical species i in mg [veh-mile]$^{-1}$
L : tunnel length in m
M_i : mass of a chemical species i emissions in mg
N : traffic count in number of vehicles per unit time

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