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NON-METHANE HYDROCARBON AND ALDEHYDE MEASUREMENTS IN BUDAPEST, HUNGARY

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Abstract—During three summer measuring campaigns the atmospheric concentration of non-methane hydrocarbons and aldehydes were measured at two sites in Budapest. Two hundred and forty-five flask samples were analyzed for non-methane hydrocarbon concentration and hydrocarbon composition. For formaldehyde and acetaldehyde concentration 185 and 122 samples were analyzed, respectively. The total non-methane hydrocarbon concentration shows a characteristic diurnal variation with a peak between 6 a.m. and 9 a.m. At the two sites the average concentrations between 6 a.m. and 9 a.m. are 802 and 606 ppbC, respectively. Comparing the speciation of hydrocarbons in the air of Budapest with that measured in other cities we have realized a relative surplus in C_6 alkanes which is balanced by the low contribution of C_3–C_4 alkanes.

Both the formaldehyde and acetaldehyde concentration were found rather high. In the downtown the average concentraations are 10.4 and 4.4 ppb, while 3 km away from the center of the city the corresponding values are 28.0 and 5.8 ppb.

Key word index: Urban measurements, non-methane hydrocarbons, aldehydes.

INTRODUCTION

Non-methane hydrocarbons (NMHC) play an important role in photochemical air pollution as precursors of oxidants. Since the late 1960s NMHC concentrations have been reported from several urban areas (see, e.g. Stephens and Burleson, 1969; Lonneman *et al.*, 1974; Nelson and Quigley, 1982; Wathne, 1983; Grosjean and Fung, 1984; Sexton and Westberg, 1984; Colbeck and Harrison, 1985; Singh *et al.*, 1985; EPA, 1986; Puxbaum and Lanzerstorfer, 1986; Jones, 1988; Rao and Pandit, 1988), however, there are hardly any data from Eastern European cities (Isidorov *et al.*, 1983). The bibliography of the papers reporting urban aldehyde concentration data is not as long as that of NMHC data but is till impressive (see, e.g. Grosjean, 1982; Grosjean and Fung, 1984; Schulam *et al.*, 1985; Nantke and Lohrer, 1986; Salas and Singh, 1986; Kalabokas *et al.*, 1988), however, we could not find any data from Eastern European cities.

One of the primary sources of hydrocarbons in urban areas is automobile traffic involving petrol distribution, evaporation and automobile exhaust (e.g. Derwent and Hov, 1979; Hough and Derwent, 1987; Rutten *et al.*, 1988; Molnár, 1990). Aldehydes are produced directly in the combustion of hydrocarbon fuels and indirectly in the atmospheric photooxidation of the same fuels (Derwent and Hov, 1979; Baugh *et al.*, 1987; Calvert and Madronich, 1987). From 1970 until 1989 the number of cars in Budapest (525 km^2, 2 million inhabitants) increased from 82,000 to 448,000 (KSH, 1989). It has caused a dramatic increase in the traffic-originated air pollution. The composition of the car park in Budapest (and in some other Eastern European cities) is rather special because one third of cars have two-stroke engines which emit much more hydrocarbons and less nitrogen oxide than a four-stroke engine (Merétei and Borsi, 1990). If it causes a significant difference in the composition of volatile organic compounds, it should be taken into account in air pollution modelling, not only on a local scale but—because of the quantity of air pollutants emitted—probably on a regional scale as well. However, while for NO_x, CO and SO_2 a monitoring network has been established in Budapest, for hydrocarbons no measurements were performed until 1987.

In summer 1987, 1988 and 1989 in the frame of a photochemical air pollution study the Institute for Atmospheric Physics and the Central Research Institute for Chemistry, both in Budapest, measured the atmospheric concentration of non-methane hydrocarbons, as well as the concentration of aldehydes. Two stationary measuring sites were set up for the study: Site 1 in downtown Budapest, while Site 2 at the border of the downtown, approximately 3 km away from the center of the city (Fig. 1). Both measuring platforms were located at rooftop level (approx. 20 m above street level), above the top of the neighboring

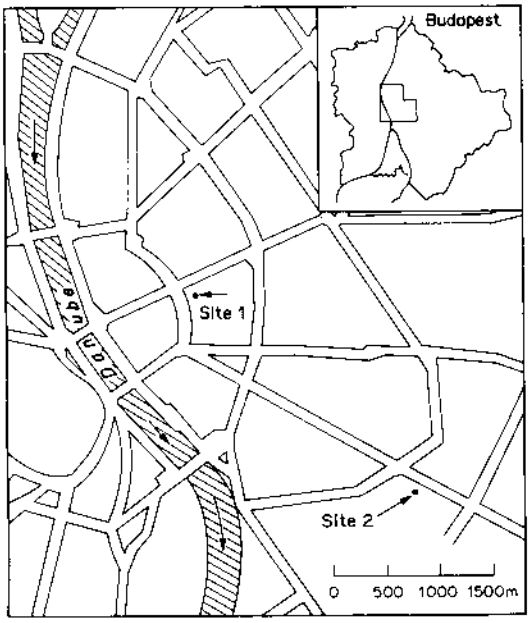

Fig. 1. The locations of the measuring sites.

buildings to increase the spatial representativeness of the measurements. As the primary aim of the study was the investigation of the photochemical air pollution, all the samples were taken on calm, sunny working days.

EXPERIMENTAL

The air samples for hydrocarbon analysis were taken in evacuated (10^{-2} Pa) Pyrex glass bulbs of 1 or 2 ℓ equipped with Teflon valves. The analysis was performed by gas chromatography after a preconcentration procedure which was based on the method elaborated by Schmidbauer and Oehme (1985). A measured volume of the sample was drawn through a drying tube filled with granular magnesium perchlorate held at 60°C and a stainless steel spiral tube (1/8 in o.d., filled with quartz chips) cooled by liquid nitrogen. All hydrocarbon components trapped in the spiral were cryofocussed into the first section of the chromatographic capillary column and then injected into the column by heating this section with hot water. For the separation a 30 m long Pyrex glass capillary column of 0.25 mm internal diameter was used. The stationary phase was chemically bonded OV-1/OV-101 mixed silicon polymer of 3–4 μm thickness. Hydrogen was used as carrier gas. A HP5840A gas chromatograph equipped with FID was applied for the temperature programmed analysis (start at −5°C, after 5 min the temperature was increased up to 135°C using a heating rate of 5°C min^{-1}).

Hydrocarbon species were identified by retention times of authentic samples. For quantification multicomponent mixtures of known concentrations were prepared.

Detection limit, defined as the smallest recognizable peak on the chromatogram, was approximately 1.5 ppbC for all the components. The total non-methane hydrocarbon (TNMHC) concentration was determined from the sum of peak areas using a calibration factor determined experimentally.

Our GC column could separate ethene–ethine–ethane in synthetic mixtures, however it was unable to resolve them properly in ambient air samples, perhaps as a consequence of water remained in the samples. This causes some uncertainty in ethene concentrations measured. We could not identify each component in the most complex part of the chromatogram, but it was possible to classify them into groups according to their retention times. For example, the sum of unidentified components having retention times between that of *n*-hexane and *n*-heptane (on our apolar column) is given as (C_6–C_7) in Table 1. Such a group consists of species of different chemical nature (aliphatic and olefinic isomers) but represents a definite molecular weight range.

The loss of hydrocarbons was carefully examined in each stage of the analysis (i.e. in sampling, drying, concentrating, focusing and GC separation) as well as in the whole procedure. Using synthetic hydrocarbon mixtures of known composition it was found that at least 95% of non-methane hydrocarbons could be recovered from an air sample.

Repeated analyses performed on both synthetic and ambient samples in ppbC range indicated that there is no measurable change in hydrocarbon concentrations within 1–2 days of storage. Only the concentration of the highest molecular weight components (like xylenes and C_{10} aliphatics) showed a certain decrease during longer storage periods. Cross-contamination between successive samples was not observed.

For the measurement of aldehyde concentrations, the air was bubbled through the solution of 2,4-dinitrophenylhydrazine in 2N HCl. The sampling efficiency of this method is good for formaldehyde but moderate for acetaldehyde (Grosjean and Fung, 1982). The sampling time was 1 h with 30 ℓh^{-1} air flow. The phenylhydrazone derivatives were extracted with CCl_4. After concentration, the samples were analyzed by GC with a flame ionization detector (see, e.g. Rutten *et al.*, 1988). For the separation, a 30 m long DB1 Durabond fused silica capillary column of 0.25 mm internal diameter was used. 3-Pentanone was applied as an internal standard. For calibration, standard solutions were used.

Parallel samples were taken for formaldehyde determination by chromotropic acid method (APHA, 1977; Fushimi and Miyake, 1980), which is a simple method with negligible interferences. This is the national standard method for formaldehyde measurements in Hungary (MSZ 21456/10–84). The volume of the sampling solution was 5 cm^3 and the air flow was adjusted to 30 ℓh^{-1}. The sampling efficiency of the method is above 90%, and the detection limit at the conditions we used was approximately 10 ppb. The project offered a posibility to compare the chromotropic acid method with the GC version of the DNPH method. The HCHO concentration measured by the two methods are in acceptable agreement. The linear regression analysis gives a correlation coefficient of 0.85. The intercept (-0.67 ± 2.16) does not differ significantly from zero. The slope is 0.84 ± 0.06. On the basis of this comparison we believe that the relatively old-fashioned chromotropic acid method is still an acceptable procedure. Salas and Singh (1986) obtained a similar conclusion comparing the chromotropic acid method with the HPLC version of the DNPH procedure.

As a quality control the formaldehyde concentrations obtained by the two methods were regularly compared. If the differences was not bigger than 50%, the average of the two values was accepted as the atmospheric concentration. Otherwise we relied on the chromotropic acid method. The reason for this was the fact that in some cases we had problems with the DNPH method due to the non-satisfactory purity of the sampling solution. Because of this uncertainty and the uncertain sampling efficiency discovered by Grosjean and Fung (1982), the acetaldehyde concentration data can be considered only as approximate values.

Table 1. Non-methane hydrocarbon composition between 6 a.m. and 9 a.m. in Budapest (ppbC). Blank as the lower end of the range means that there was at least one sample in which the concentration was below the detection limit

Species	Site 1 Range	Site 1 Arithmetic mean	Site 1 Standard deviation	Site 1 Geometric mean	Site 2 Range	Site 2 Arithmetic mean	Site 2 Standard deviation	Site 2 Geometric mean
Ethylene	14.3–287.0	68.0	39.2	59.9	8.6–150.8	50.0	32.8	40.3
Ethane	5.2–35.0	15.6	6.4	14.3	5.5–136.9	27.2	23.9	20.6
Propene	–95.6	11.1	13.4	6.2	–37.0	8.4	8.4	4.3
Propane	1.8–29.3	10.4	5.6	8.8	1.4–71.3	16.2	15.0	11.1
1– + *i*-Butene	–37.7	7.8	7.2	4.5	–35.0	5.3	6.5	2.4
2-Butene	–25.5	5.0	4.8	2.9	–16.0	2.7	3.7	1.3
i-Butane	–30.1	12.4	6.4	10.5	1.0–36.0	10.7	7.8	7.8
n-Butane	4.0–55.3	19.7	10.3	17.1	1.7–66.4	17.7	11.5	14.2
i-Pentane	13.5–102.3	51.5	21.3	46.5	5.0–138.0	39.3	25.8	31.8
n-Pentane	10.0–58.3	27.2	11.1	25.0	4.8–110.0	25.2	19.6	20.1
2-Methyl-pentane	7.8–52.9	28.4	11.7	25.7	1.7–80.0	22.4	15.3	17.9
3-Methyl-pentane	5.3–41.1	19.0	8.1	17.1	–52.0	15.2	10.0	11.9
(C_5–C_6)	–65.0	9.8	16.5	1.9	–118.0	23.0	28.1	5.9
n-Hexane	9.1–82.3	34.4	13.4	31.8	4.9–82.0	25.0	14.0	21.6
(C_6–C_7)	–103.6	21.9	33.6	2.6	–193.0	40.4	45.8	9.9
n-Heptane	3.7–31.9	15.0	6.9	13.3	–36.0	11.3	7.7	8.9
(C_7–C_8)	–91.7	11.8	19.7	2.0	–82.0	13.7	18.8	3.8
n-Octane	1.8–32.4	7.5	4.5	6.5	–18.0	4.8	3.6	3.6
(C_8–C_9)	–53.6	5.6	11.3	1.3	–75.0	8.5	14.1	2.1
n-Nonane	1.5–34.1	8.0	5.4	6.6	–15.4	4.7	3.1	3.8
(C_9–C_{10})	–215.1	25.4	46.4	2.6	–149.0	26.7	34.1	6.2
n-Decane	2.1–43.7	13.4	8.4	11.2	–46.0	7.2	6.0	5.3
(>C_{10})	–156.1	15.7	31.2	2.1	–114.0	15.4	24.3	3.7
Benzene	6.9–55.3	27.4	11.8	24.8	4.2–158.6	25.8	23.0	19.9
Toluene	8.4–124.6	60.8	30.4	52.2	9.2–147.2	46.8	28.5	39.1
Ethylbenzene	1.6–45.5	14.5	8.3	12.2	–28.0	10.0	5.9	8.0
m/*p*-Xylene	2.5–169.6	42.7	30.1	34.0	1.7–82.9	28.6	16.8	23.6
o-Xylene	–66.4	14.1	11.0	10.9	–30.0	9.7	6.1	7.7
TNMHC	180.0–1726.0	802.0	370.3	708.9	96.0–1750.0	605.9	374.9	497.2

HYDROCARBONS

During the periods of 17 August–2 September 1987, 5 July–18 August 1988, as well as 10–25 July 1989, 245 samples were taken for hydrocarbon analysis.

The majority of the samples (171) were taken between 6 a.m. and 9 a.m., i.e. during the morning peak of air pollution. The results are summarized in Table 1. As the data were distributed lognormally rather than normally (Fig. 2), the geometric means are also listed.

The diurnal variations of the total non-methane hydrocarbon concentration at the measuring sites show similar patterns (Fig. 3). At Site 1 (downtown), the morning peak is somewhat higher and wider towards late morning than at Site 2, which can be explained by the different traffic conditions. However, there is no significant correlation between the individual concentration values measured at Site 1 and Site 2 at the same time. The main reason for this is the different location of the stations. The emission field is roughly isotropic around Site 1; however, this cannot be said about Site 2. Site 2 obtains slightly polluted air from the sector of SE–W. The downtown, with a high emission density is in the northwesterly direction, and there is a main street with remarkable traffic in the northeasterly direction. Thus the wind direction is a main factor controlling the hydrocarbon level at Site 2, while it hardly influences the concentrations at Site 1.

Mixing height and wind speed also seem to be important controlling factors. Unfortunately, the present data base is not sufficient for a multivariate statistical analysis of hydrocarbon concentrations and meteorological parameters.

Although, during the last years, several papers have been published on hydrocarbon measurements (see Introduction), the direct comparison of the data is rather problematic. The significant diurnal variation in the emission as well as in the intensity of the atmospheric dilution makes the time of sampling critical. The height of the sampling site (Puxbaum and Lanzerstorfer, 1986) and the region of sampling (downtown, suburb, industrial region, highway, etc.) can also influence the concentrations measured. In addition the transformations initiated by the photochemical processes may modify the hydrocarbon composition during the day.

It is believed that the most similar sampling conditions to those in Budapest were during the programs published in a paper by Sexton and Westberg (1984) as well as in an EPA report (EPA, 1986). On the basis of these data, it seems the total non-methane hydrocarbon concentration in the air of Budapest is roughly as high as, or even higher than, that in the air of the big American cities monitored.

At both sites in Budapest one-third of TNMHC is alkane on the ppbC base. Ten per cent are alkene and 20% are aromatics. The rest of TNMHC is in the $C_n - C_{n+1}$ groups, which may involve both alkanes and alkenes, or unidentified. Unfortunately, these values are incomparable with data from other cities (e.g. Isidorov *et al.*, 1983; Sexton and Westberg, 1984) because different authors measured different hydrocarbon components and TNMHC is also defined in different ways.

Most of the hydrocarbons measured have a lifetime of a couple of hours (Uno *et al.*, 1985) which makes the hydrocarbon composition less changeable with time than the concentrations themselves. Therefore, in the comparison of the speciation, we may take less care with the similarity of the sampling conditions. Alkanes and aromatics are studied separately because a part of the authors measured only the first or the second group. Alkenes are left out from the comparison. In our experiment only few alkene species were measured from which the concentration of the butene isomers were often below the detection limit and the ethene measurements also involve uncertainties as mentioned in the Experimental section.

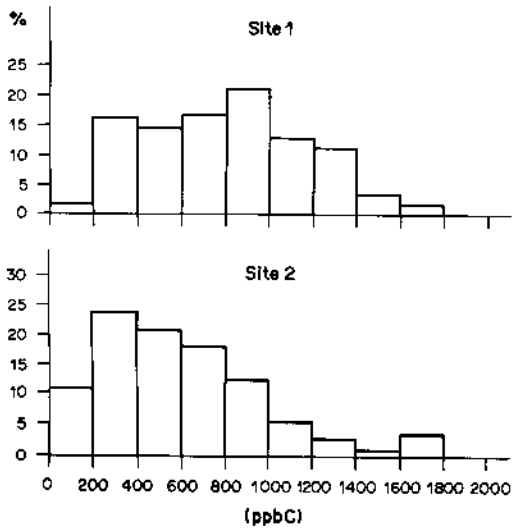

Fig. 2. Frequency distribution of total non-methane concentration.

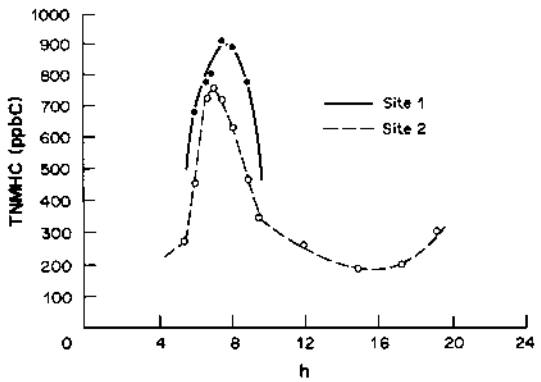

Fig. 3. Average diurnal variation of total non-methane hydrocarbon concentration. Dots represent the average concentration of samples taken in the same hours of the days.

For the comparison those alkane and aromatic species have been chosen which were measured in most experiments. The relative contributions of each species in ppbC base to the total of the chosen components have been calculated and are presented in Tables 2 and 3.

In the case of alkanes we find lower propane and butane ratios and a higher *n*-hexane one than in the other cities listed. There is also a slight difference in the composition between the two measuring sites in Budapest.

In the case of aromatics the same composition was observed at both sites in Budapest. The values are in good agreement with most measurements from other cities. Only the compositions reported by Sexton and Westberg (1984) deviate from them a little: the toluene/benzene ratio is lower in Budapest. Isidorov and his coworkers' (1983) data are not presented in Table 3 because they did not measure the concentration of ethylbenzene. If we compare only those four aromatics which were measured in both experiments we find that the toluene/benzene ratio is significantly lower in Leningrad than in Budapest.

The differences may be explained by the different emission in the cities. These cities may differ in population, industry, automobile traffic, climate (e.g. gasoline

Table 2. Percentage distribution of some alkanes on ppbC base in different cities

Species	Budapest (1) Site 1	Budapest (1) Site 2	Harwell (2)	Lancaster (3)	Sydney (4)	Tokyo (5)	Seven American cities (6)
Ethane	10	17	19	26	10	9	8–12
Propane	6	9	13	6	12	25	9–17
i-Butane	7	6	17	23	13	9	10–13
n-Butane	11	11	26	7	21	16	24–33
n-Pentane	17	16	7	5	17	13	14–19
2-Methylpentane	17	14	6	9	11	9	8–11
3-Methylpentane	11	10	5	19	7	4	5–8
n-Hexane	21	17	7	5	9	15	6–8

Species	Budapest (1) Site 1	Budapest (1) Site 2	Vienna (7)	Leningrad (8)
n-Butane	15	18	19	23
n-Pentane	22	26	24	26
n-Hexane	29	28	23	23
n-Heptane	12	11	17	14
n-Octane	6	5	5	7
n-Nonane	6	5	4	4
n-Decane	10	7	8	3

(1) Present study.
(2) Jones (1988).
(3) Colbeck and Harrison (1985).
(4) Nelson and Quigley (1982).
(5) Uno *et al.* (1985).
(6) Sexton and Westberg (1984).
(7) Puxbaum and Lanzerstorfer (1986).
(8) Isidorov *et al.* (1983).

Table 3. Percentage distribution of some aromatics on ppbC base in different cities

Species	Budapest (1) Site 1 and Site 2	Harwell (2)	Tokyo (3)	Vienna (4)	Seven American cities (5)	Fifteen American cities (6)
Benzene	20	18	13	20	9–19	17–27 (Pittsburgh: 36)
Toluene	39	37	49	39	35–50	31–45
Ethylbenzene	8	11	13	8	7–11	7–15
m, p-Xylene	25	21	17	23	20–28	15–25
o-Xylene	8	13	8	10	7–13	6–15

(1) Present study.
(2) Jones (1988).
(3) Uno *et al.* (1985).
(4) Puxbaum and Lanzerstorfer (1986).
(5) Sexton and Westberg (1984).
(6) Singh *et al.* (1985).

Table 4. Aldehyde concentrations in Budapest (ppb)

	HCHO				CH_3CHO			
	Range	Arithmetic mean	Standard deviation	Geometric mean	Range	Arithmetic mean	Standard deviation	Geometric mean
Site 1	–58.0	14.9	12.8	10.4	–27.0	6.5	5.9	4.4
Site 2	7.0–176.0	34.6	25.8	28.0	–35.0	8.9	8.2	5.8

Blank as the lower end of the range means that there was at least one sample in which the concentration was below the detection limit.

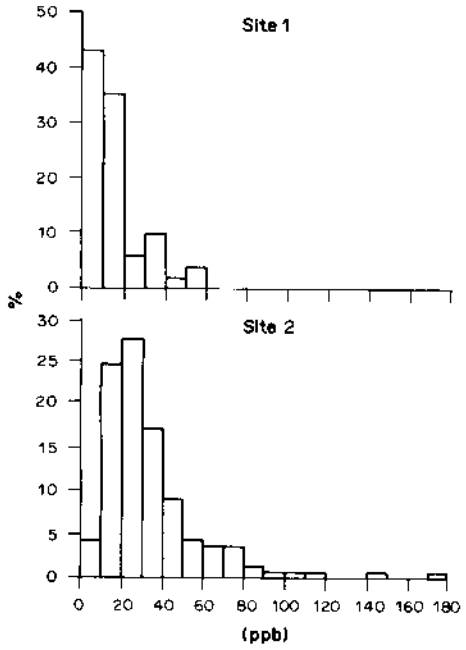

Fig. 4. Frequency distribution of formaldehyde concentration.

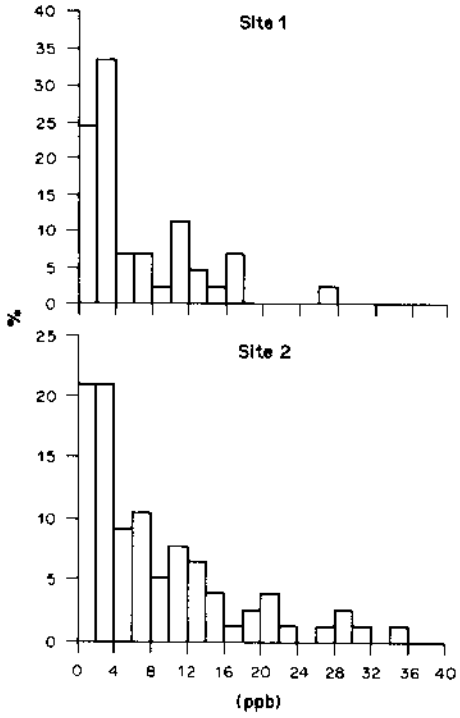

Fig. 5. Frequency distribution of acetaldehyde concentration.

evaporation), gasoline and car park composition (see e.g. Pandit and Rao, 1990), emission control regulations and many other influencing factors. The differences and similarities could be explained only in that case if emission inventories and other statistical data were available for all the cities compared.

ALDEHYDES

During the three summer measuring campaigns 185 chromotropic acid and 122 DNPH samples were taken to determine the concentration of aldehydes. The results are summarized in Table 4, while the frequency distributions are presented in Figs 4 and 5.

The formaldehyde concentration at Site 2 is significantly higher than at Site 1. At Site 2 some extremely high values (100–176 ppb) were also measured. The relative absence of low concentrations in the frequency distribution (see Fig. 4) also suggests a direct, more or less permanent local HCHO emission.

Perhaps because of the reason mentioned above the formaldehyde concentration at Site 2 does not show any characteristic diurnal variation (Fig. 6a). The HCHO concentration at Site 1 gradually decreases during the day. Comparing this daily pattern with that of total non-methane hydrocarbon concentration we find that the more intensive dilution during the afternoon is largely compensated by the photochemical production of HCHO. (We assume that there is no formaldehyde source which operates mainly in the afternoon.)

The formaldehyde concentrations at Site 1 are similar to those in the American cities presented by Salas and Singh (1986), as well as by Schulam *et al.* (1985) and only a few exceed the values measured around Paris (Kalabokas *et al.*, 1988). The concentrations at Site 2 are higher than those published in the literature and they frequently exceed the maximum

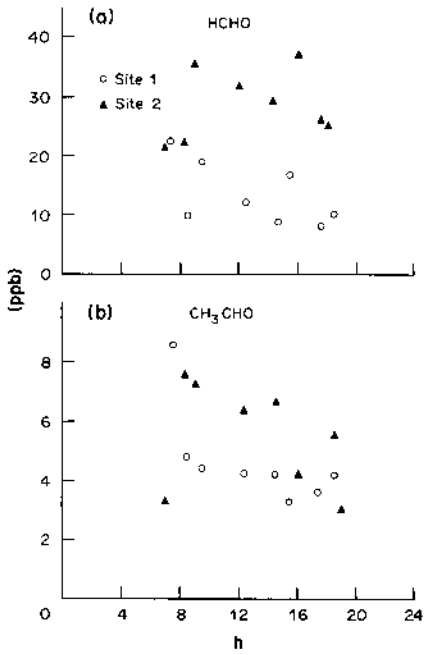

Fig. 6. Diurnal variation of formaldehyde (a) and acetaldehyde (b) concentration.

value allowed by the Hungarian Health Standard ($35\ \mu g\ m^{-3}$ = approx. 28 ppb; 30 min value).

The acetaldehyde concentrations at Site 2 are also somewhat higher than at Site 1 but they show a similar diurnal pattern (Fig. 6b). At both sites the concentration decreases during the day. Comparing the data with those published in the literature (Schulam *et al.*, 1985; Salas and Singh, 1986; Kalabokas *et al.*, 1988), the acetaldehyde concentrations measured in Budapest seem rather high. The concentration frequently, sometimes significantly, exceeds the Hungarian Standard ($10\ \mu g\ m^{-3}$ = approx. 5.4 ppb; 30 min value).

A possible reason for the high aldehyde concentration in Budapest may be the high number of cars with two-stroke engines (Merétei and Borsi, 1990). However, unfortunately, this idea cannot be supported satisfactorily yet because of the limited number of data for the composition of the exhaust gas of two-stroke engines typical in Budapest. The higher aldehyde concentration at Site 2 may be explained by unknown local sources, perhaps by the hospitals and clinics in the vicinity of the sampling site.

CONCLUSIONS

On the basis of the analysis of air samples we could obtain an impression of the hydrocarbon and aldehyde pollution in the air of Budapest. We have found that the concentrations of these organics are as high as, or in some cases even higher than, the big cities of the world reported in the literature. We have discovered certain deviations in the speciation of hydrocarbons between Budapest and the other cities in the comparison and we have also realized that the aldehyde concentrations are rather high in Budapest. However, in the lack of emission inventories and reliable data for the exhaust gas composition of two-stroke engines, these differences cannot be definitely attributed to the special car park composition in Budapest at the present phase of the research. It may be simply the consequence of different gasoline composition.

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