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Mono- and di-carboxylic acids under long-range transport of air pollution in central Japan

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ABSTRACT

Behavior of mono- and di-carboxylic acids during the long-range transport of photochemical air pollution from the metropolitan, large emission source area along the Tokyo Bay to the inland mountainous region in central Japan was investigated as a part of a cooperative field observation of meteorology and chemistry in summers, 1983 and 1986. Measurements were made for every 3 h at inland sites along the transport route of the polluted air mass. Concentrations of acetic acid, propionic acid and *n*-butyric acid increased to the ranges of 4–8 ppb (V/V), 0.3–0.7 ppb and 0.1–0.3 ppb, respectively in the daytime. In particular, during day, they increased significantly when the polluted air mass arrived at the sampling sites, while they decreased at night. These diurnal variations were similar to that of ozone. Of the di-carboxylic acids, succinic acid was the most abundant species, followed by phthalic acid and malonic acid. Concentrations ranged from 20–130 ng m^{-3} in the daytime, and were much higher in the daytime than at night. These diurnal variations were similar to those of ozone. These facts suggest that most of mono- and di-carboxylic acids were produced by photo-oxidation of anthropogenic compounds during long-range transport.

1. Introduction

Mono- and di-carboxylic acids are important to understand the fate of the airborne organic species. By photochemical reactions of the gaseous organic pollutants in the atmosphere oxidants and various binds of radicals are formed, which in turn, oxidize the organic species to produce mono- and di-carboxylic acids. It is suggested in smog chamber by Pitts et al. (1977), Akimoto et al. (1980), Grosjean (1977), Grosjean and Friedlander (1980) and Hatakeyama et al. (1985 and 1987). However, few volatile mono-carboxylic acids except for formic acid have been studied in the atmosphere. Dawson et al. (1980), Hoshika (1982), and Kawamura et al. (1985) reported C_1–C_2, C_2–C_6 and C_1–C_{10} mono-carboxylic acids, respectively, but these studies have been made only for monoacids.

Grosjean et al. (1978) reported that C_3–C_9 aliphatic di-carboxylic acids were produced significantly in the form of aerosol during a smog episode in the Los Angeles area. Kawamura and Kaplan (1987) detected C_2–C_{10} aliphatic diacids, including unsaturated, and phthalic acids in the Los Angeles area. Yokouchi and Ambe (1986) detected C_2–C_{16} aliphatic diacids and phthalic acids in the mildly polluted atmosphere in Tsukuba, Japan. The sampling times in those measurements were long and the time variations difficult to understand fully. The purpose of the present investigation is to make clear the behavior of mono- and di-carboxylic acids in severely polluted air masses.

In summer in central Japan, long-range transport of photochemical air pollution occurs very frequently. A large-scale wind field is induced by the local winds and transports polluted air formed near the Tokyo metropolitan area 200 km inland into the mountainous region (Kurita et al., 1985; Kurita and Ueda, 1986a and 1986b).

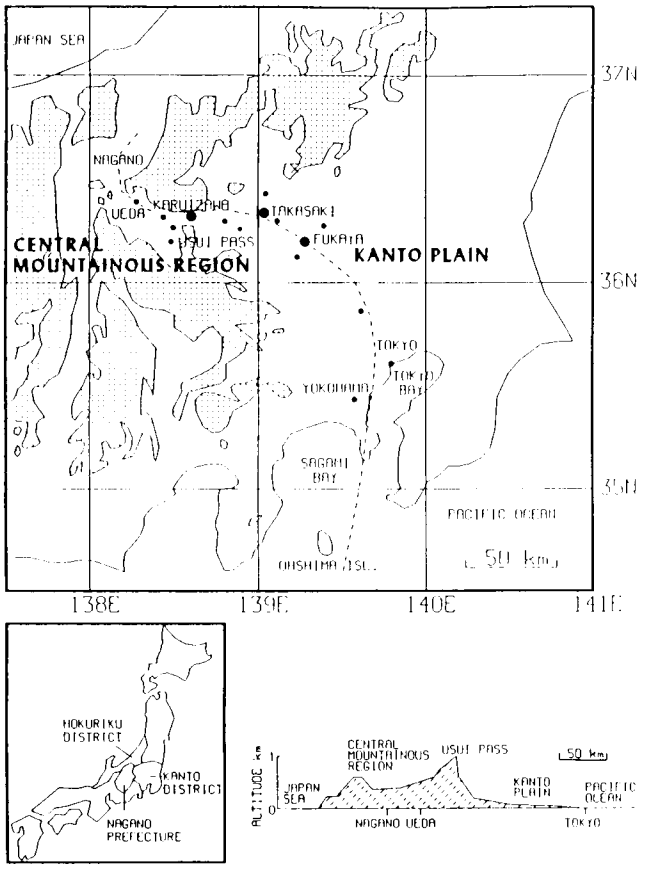


Fig. 1. Geographical features and sampling sites. The dotted line represents the actual route of the long range transport. Land higher than 1000 m above MSL is denoted by the stippled areas.

During the long-range transport, nitrogen oxides (NO_x) and hydrocarbons react photochemically to produce secondary pollutants such as ozone, nitric acid and nitrates (Satsumabayashi et al., 1986; Kurita and Ueda, 1986b; Sasaki et al., 1988). As a result, high concentrations of ozone and suspended particulate matter are frequently observed in the mountainous region. Moreover, the daily maximum concentration occurs in the late evening. For example, in 1979, the city of Ueda (Fig. 1) ranked 7th in Japan in terms of days with ozone above 120 ppb (V/V), even though it ranked beyond 150th in terms of population size.

In this paper, the behavior of mono- and dicarboxylic acids was investigated in relation to the photochemical reactions during the long-range transport from the large emission source area along the Tokyo Bay to the inland mountainous region. The diurnal variations and composition of the acids are studied.

2. Geographic features and long-range transport

The measurements were made a cooperative field research of meteorology and chemistry was made in summers of 1983 and 1986. The region under study is located in central Japan. Geographic features and the actual path of the long-range transport are shown in Fig. 1. The Kanto Plain, facing the Pacific Ocean is the largest plain (15,000 km^2) in Japan. The mountainous region consists of mountain chains 2–3 km high with many basins and valleys that form a

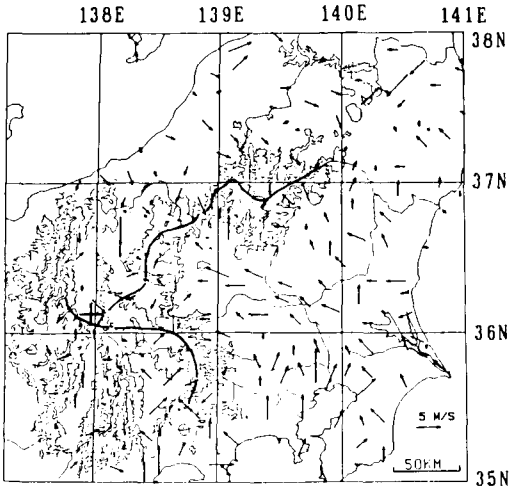


Fig. 2. Surface wind field at 1500 JST on 29 July 1983. The thick solid line represents a convergence line between the large-scale winds from the Pacific Ocean and the Japan Sea, and the cross represents a thermal low center.

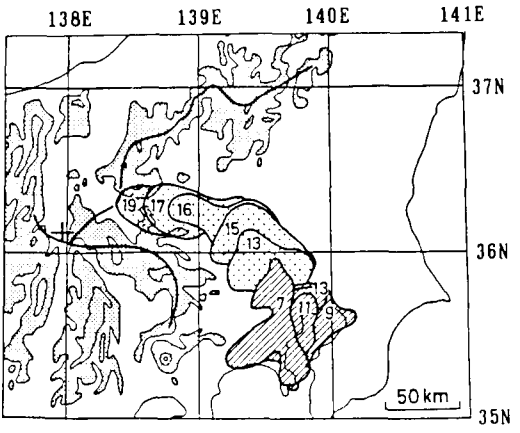


Fig. 3. Movement of high-concentration zones of ozone and NO_x on 29 July 1983. Stippled areas: ozone concentration > 100 ppb; hatched areas: NO_x concentration > 40 ppb. Numbers indicate the time (hour).

complex terrain. The average height of this region is 1200 m above MSL. Those regions are connected by the Usui Pass, about 1000 m above MSL. Large emission sources are concentrated in the populated and industrialized area along the Tokyo Bay.

Long-range transport of photochemical air pollution occurs very frequently on clear summer days from the metropolitan, large emission source area along the Tokyo Bay to the inland mountainous region. It is driven by a large-scale wind system, which is induced thermally and locally by a combination of local winds under light gradient wind conditions. That is, in the daytime a land/sea breeze and steady onshore wind develop intensely to create an extended sea breeze in the Kanto Plain. On the other hand, in the mountainous region a strong thermal low is generated (the atmospheric pressure is by 4–6 mb less than the coastal region) and sustained until late evening. Then, a wind blowing into the thermal low combines the extended sea breeze and upslope winds, etc. into a large-scale wind system (Fig. 2) and a long-range transport occurs in this large-scale wind field. Thus, the long-range transport of this type is the most typical transport pattern in summer in central Japan and it takes almost a fixed route determined by the topography.

Fig. 3 shows the movement of high concentration zones of ozone and NO_x on 29 July 1983, when the long-range transport occurred. A highly polluted air mass was created by land breezes and covered the Tokyo Bay in the previous night. It remained there in the morning calm. At 1200 JST, when the sea breeze started, the polluted air mass was transported inland and created the high concentrations of ozone. At 1500 JST, the wind was blowing into the thermal low pressure (Fig. 2). At this time, the high concentration zone of ozone reached to Fukaya. This wind field continued unchanged until 1900 JST and drove the polluted air mass farther inland. Finally, the high concentration zone of ozone reached Karuizawa in the mountainous region at 1800 JST, after travelling 150 km from the main sources.

3. Experimental studies on mono- and di-carboxylic acids

Measurements of mono- and di-carboxylic acids were made as a part of the cooperative field research of meteorology and chemistry. Sampling sites were established at Fukaya, Takasaki and Karuizawa along the path of the polluted air mass but far from the large emission sources. As shown in Fig. 1, Fukaya and Takasaki are in the Kanto Plain, 70 km and 85 km, respectively,

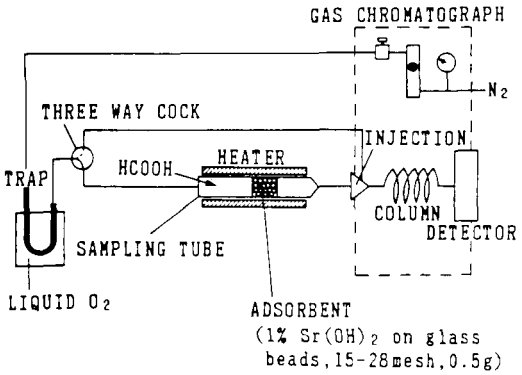


Fig. 4. Schematic diagram of mono-carboxylic acid analysis.

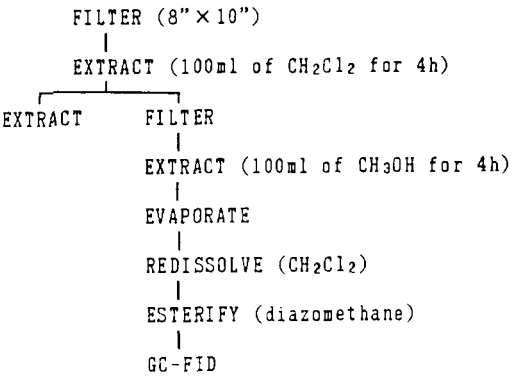


Fig. 5. Flow sheet of di-carboxylic acid analysis.

inland from the large emission source area. Karuizawa is just near the mountain pass, bounding the Kanto Plain and the mountainous region. Including those sites, there were 7 intensive pollutant monitoring sites in total which measured NO_x, ozone, SO_2, sulfate, nitrate, HNO_3, TSP (total suspended particle), NH_4^+ and 30 hydrocarbon species.

Mono-carboxylic acids were present mostly in vapor phase (Kawamura et al., 1985). They were collected on 0.5 g of glass beads (15–28 mesh) coated with 1 wt% strontium hydroxide and packed in a sampling tube (5 mm i.d. and 180 mm long). The sampling flow rate was set at 2 l min^{-1}. The sampling time and interval were three hours. Analysis of the mono-carboxylic acids was made, following Hoshika's method (1982). As shown in Fig. 4, after the sampling tube was connected to gas chromatograph (Shimadzu GC-5A), the trapped monoacids were desorbed with 5 μl of formic acid at 200°C under nitrogen carrier gas flow at a flow rate of 50 ml min^{-1} and detected with a flame ionization detector. The analytical column was a glass column (3 mm i.d. and 1.5 m long), packed with 0.3% FFAP + 0.3% H_3PO_4 on Carbopack B (60–80 mesh). The column temperature was increased from 110°C to 240°C at a rate of 8°C min^{-1}.

Sampling efficiencies of the mono-carboxylic acids were more than 84%. Precisions (as CV%, $n = 5$) were less than 10%. In the case of a 360 l sample gas volume, detectable limits were about 0.005 ppb. Blank values were less than 5% of the sample peaks. Although aldehydes were considered to be oxidated to acids during the sampling collection, it was assumed negligible. It was confirmed for acetic acid, that is, even when acetaldehyde added to the polluted air to 40 ppb in a tedlar bag was passed through the sampling tube, the concentration of the acetic acid resulted in the same value as that without addition.

Of the di-carboxylic acids, oxalic acid is the most abundant (Kawamura and Kaplan, 1987). It is mostly in the vapor phase, whereas other di-carboxylic acids are mostly in the form of aerosol. Here di-carboxylic acids in the aerosol phase were collected on a quartz fiber filter (Pallflex 8″ × 10″) using a high-volume air sampler with a capacity of 1.2 m^3 min^{-1}. The sampling interval was also three hours. Prior to sampling, filters were heated at 500°C for three hours in nitrogen. Determination of di-carboxylic acids was made by the method of Yokouchi and Ambe (1986) (Fig. 5). The filter was extracted with 100 ml of dichloromethane for 4 h and subsequently with 100 ml of methanol for 4 h. After filtration, the methanol extract was evaporated at a temperature of less than 40°C to dryness. The residue was redissolved with dichloromethane, and it was derivatized with diazomethane to esterify free acids. The esterified acids were analyzed by gas chromatograph (Hewlett-Packard 5840A) equipped with a methyl silicone fused capillary column (SPC-5, 0.25 mm i.d. and 30 m long) and a flame ionization detector. The column temperature was increased from 30°C to 300°C at a rate of 8°C min^{-1}. Gas chromatograph-mass spectrometer (JMS DX-300) was used to identify di-carboxylic acid derivatives.

Recovery of di-carboxylic acids were about 80% (Yokouchi and Ambe, 1986). In the case of a

216 m^3 sampling volume, detectable limits were about 0.1 ng m^{-3}. Blank values were less than 5% of the sample peaks.

4. Results and discussion

4.1. Behavior of ozone and hydrocarbons

Fig. 6 shows the diurnal variations of surface wind vectors and ozone concentration observed at the sampling sites from 28 July to 2 August 1986. Since the emissions are highest along the Tokyo Bay area and are much smaller in these inland areas, the ozone concentrations increased significantly when the transported air mass arrived at the sampling sites in the daytime. The long-range transport is clearly indicated by the delay in the ozone peak time along the transport route, especially in 28–30 July. Time lags from Fukaya to Takasaki and Karuizawa were approximately one hour and three hours, respectively, which corresponded to ones calculated by the average transport speed of 5–7 m s^{-1} in the 50–500 m layer. Air pollutants remained in the atmosphere above the nocturnal inversion layer could be entrained when the mixed layer reaches that altitude the following morning and result in a rapid increase of the ground-level ozone concentration. It was clearly seen in the morning of 31 July and 1 August in Karuizawa in Fig. 6.

During the long-range transport, reactive hydrocarbons were transformed by photochemical reactions. Decays of those hydrocarbon concentrations were shown in Fig. 7. In order to compensate the diffusion effects, these decays were presented relative to acetylene which is relatively unreactive (Whitby and Altwicker, 1978). Decay rates were higher for more reactive components, such as propylene, ethylene, *n*-pentane and isopentane. In addition, the ratios of other hydrocarbon components to acetylene also decreased. In accord with the transformation of these hydrocarbons, organic acids were presumed to be produced by photooxidation.

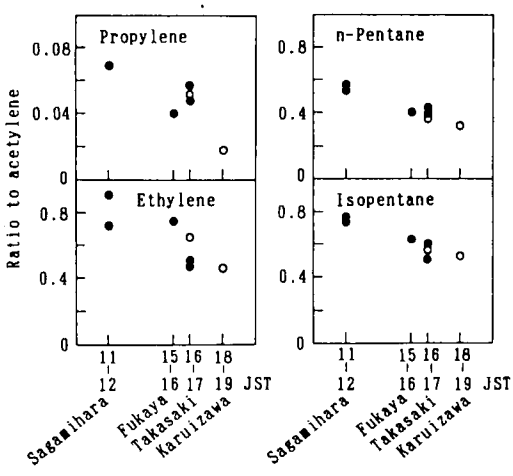


Fig. 7. Change of the ratios of hydrocarbon components to acetylene with transport time on 29 July 1983. Open circles represent the ground, and full circles the upper air (altitude 500–1300 m).

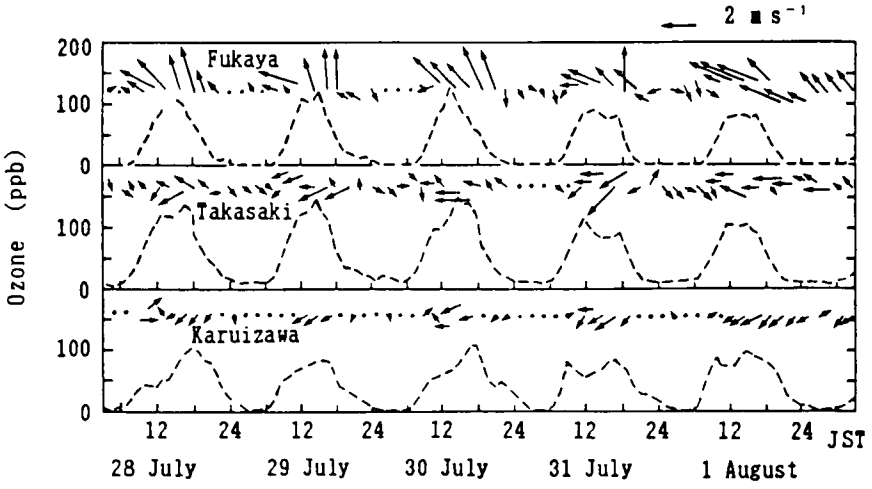


Fig. 6. Diurnal variations of surface wind vectors and ozone concentrations in 1986.

Table 1. *Averaged concentrations of mono-carboxylic acids in daytime (ppb, 0900–1800 JST, 28 July–1 August 1986)*

Acid	Fukaya	Takasaki	Karuizawa
acetic	4.71	3.96	3.48
propionic	0.39	0.44	0.27
n-butyric	0.15	0.13	0.08
isovaleric	0.01	0.01	0.00
n-valeric	0.01	0.02	0.01
n-caproic	0.00		0.01

4.2. *Mono-carboxylic acids*

Concentrations of mono-carboxylic acid components averaged over the daytime (0900–1800 JST) from 28 July to 1 August 1986 are compared in Table 1. Acetic acid was the most abundant species and its concentration attained 90% of the total mono-carboxylic acids. Concentrations of mono-carboxylic acids decrease with an increase in chain length. The values for acetic, propionic and *n*-butyric acid concentrations were in the same order of magnitude as those in Los Angeles in July and September, 1984, i.e., 1.39, 0.11, 0.031 ppb, respectively (Kawamura et al., 1985) and those in Nagoya, Japan in September and October, 1981, i.e., 1.69, 0.10, 0.025 ppb, respectively (Hoshika, 1982).

Diurnal variations of mono-carboxylic acid concentrations are shown in Fig. 8. Acetic acid, propionic acid and *n*-butyric acid increased in the daytime and decreased at night. The diurnal variations are similar among these components, and very similar to that of ozone, as seen in Fig. 9. In particular, when the polluted air mass due to the long-range transport had arrived at the observation sites and prevailed there, the concentrations were high, and higher than any reported before. This suggests that mono-carboxylic acids in the atmosphere were produced by photochemistry during the transport. On the other hand, the concentrations of valeric acid and caproic acid were below 0.02 ppb and with a diurnal variation not clearly shown in Fig. 8.

Kawamura et al. (1985) reported that motor exhaust is one of the major sources of atmospheric organic acids. Diurnal variations of SO_2, NO_x and hydrocarbon concentrations at Fukaya are compared in Fig. 10. SO_2 concentration increased in the daytime to the evening and decreased at night, while NO_x increased at night to the morning and decreased in the daytime. Since a large amount of NO_x emitted from the large emission source area along the Tokyo Bay had been transformed into nitrate and PAN during the long range transport when it arrived at the observation sites, a rather high concentration of NO_x observed in the evening was due to the local emission mainly from the mobile source. Diurnal variation of the hydrocarbon concentration was similar to that of nitrogen oxide. Thus, the high concentration of hydrocarbon in the evening was also attributed to motor exhausts near the sampling locations. However, the diurnal variations of the mono-carboxylic acid concentrations were different from those of NO_x and hydrocarbons. Therefore, it is considered that the high concentration of mono-carboxylic acids is not due to locally emitted motor exhausts but produced by photochemical reactions in the polluted air mass during transport.

4.3. *Di-carboxylic acids*

Diurnal variations of di-carboxylic acid concentrations are shown in Fig. 11. The concentrations were much higher in the daytime than those at night, and their diurnal variations were similar to that of ozone. This supports that almost all of the di-carboxylic acids were produced photochemically during the long-range transport.

Concentrations of di-carboxylic acid components averaged over the daytime (0900–1800 JST) and over all collected samples in 1986 are compared in Table 2 and Fig. 12, respectively.

Table 2. *Averaged concentrations of di-carboxylic acids in daytime (ng m^{-3}, 0900–1800 JST)*

Acid	Takasaki[1]	Karuizawa[2]
oxalic	7.8	6.2
malonic	23.3	17.7
succinic	47.1	36.4
glutaric	21.0	8.1
adipic	11.3	3.8
phthalic	38.6	31.1
terephthalic	15.3	5.9
isophthalic	3.5	1.9

[1] 27 July–31 July 1986.
[2] 29 July–31 July 1986.

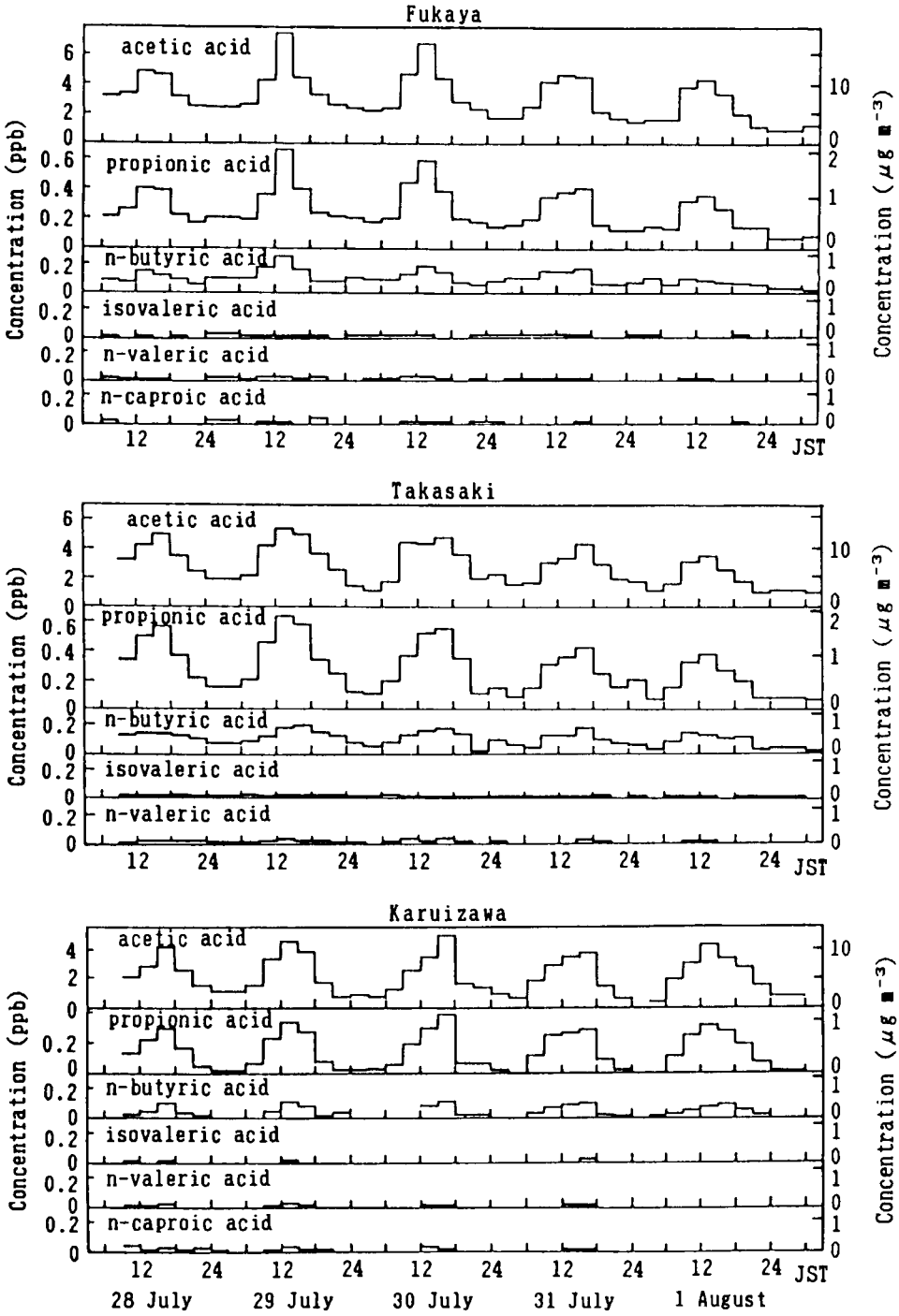


Fig. 8. Diurnal variations of mono-carboxylic acid concentrations in 1986.

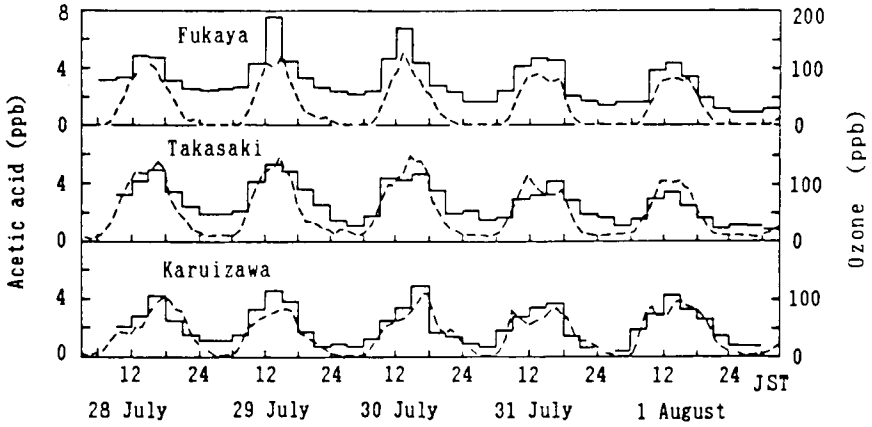


Fig. 9. Diurnal variations of ozone and acetic acid concentrations in 1986. Full lines show acetic acid, and dashed lines ozone.

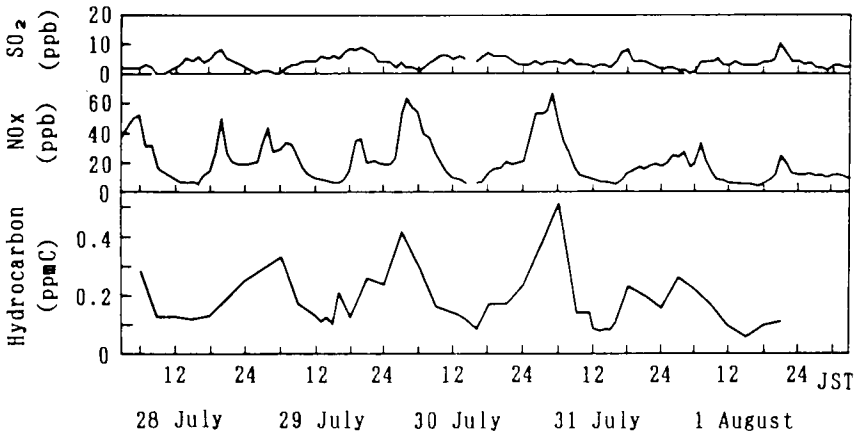


Fig. 10. Diurnal variations of SO_2, NO_x and hydrocarbon concentrations at Fukaya in 1986. Hydrocarbon concentration is the sum of the product of concentration (ppb) of 16 components by carbon number. The 16 components are ethane, propane, isobutane, *n*-butane, iso-pentane, *n*-pentane, *n*-hexane, ethylene, propylene, acetylene, benzene, toluene, ethylbenzene, *o*-xylene, *m*-xylene and *p*-xylene.

Succinic acid was the most abundant component of aliphatic di-carboxylic acids, followed by malonic acid and glutaric acid. The concentrations decreased with increasing chain length. Phthalic acid was also significantly contained in the airborne aerosols. Grosjean et al. (1978) also reported that succinic acid, glutaric acid and adipic acid were the most abundant components in the airborne aerosols during a smog episode in the Los Angeles area. However, the concentration levels observed here were from 20 to 130 ng m^{-3}, less than the values (200–500 ng m^{-3}) reported by Grosjean et al. It seems to reflect the maximum ozone concentration levels, i.e., 140 ppb in the present and 350 ppb in the study by Grosjean et al.

4.4. Formation of carboxylic acids

Herron et al. (1979) described the formation processes of mono-carboxylic acids in the gas phase. First of all, olefins such as ethylene, propylene and butene are oxidized by ozone to produce mono-carboxylic acids. Another process may be the oxidation of aldehyde by OH and

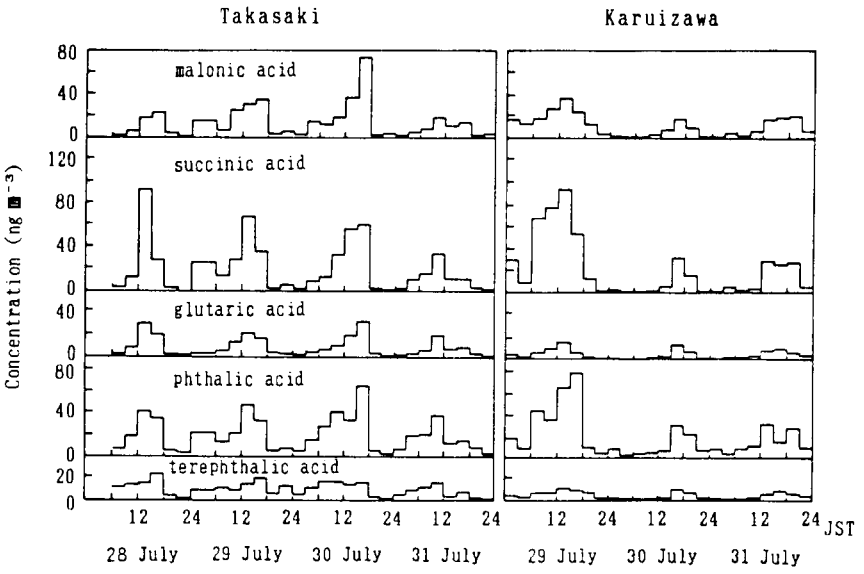


Fig. 11. Diurnal variations of di-carboxylic acid concentrations in the airborne aerosols in 1986.

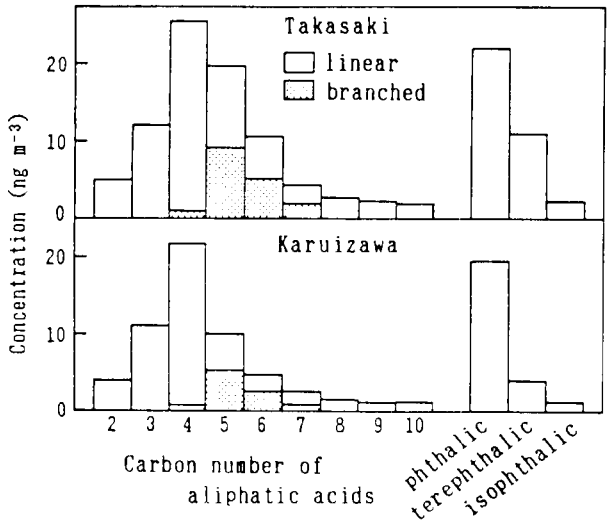
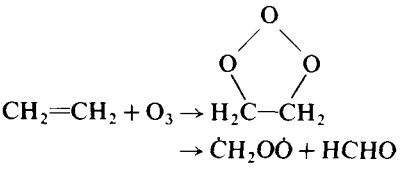


Fig. 12. Distributions of di-carboxylic acid components in the airborne aerosols, averaged for all collected samples.

HO_2 radicals. The formation of formic acid may be written as follows:

olefin + O_3 → mono-carboxylic acid



$$CH_2{=}CH_2 + O_3 \rightarrow H_2C{-}CH_2 \rightarrow \dot{C}H_2O\dot{O} + HCHO$$

$$\dot{C}H_2O\dot{O} \rightarrow CO_2 + 2H(H_2) \quad 27\% \qquad (1)$$

$$\rightarrow CO + H_2O \quad 67\% \qquad (2)$$

$$\rightarrow HCOOH \quad 6\% \qquad (3)$$

$$\dot{C}H_2O\dot{O} + HCHO, H_2O \rightarrow HCOOH \qquad (4)$$

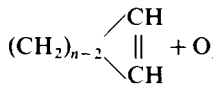
aldehyde + HO_2, OH → mono-carboxylic acid

$$HCHO + HO_2, OH \rightarrow HCOOH \qquad (5)$$

The one is the oxidation of olefin by ozone. CH_2O_2 radical and formaldehyde are produced, the former being transformed into formic acid (reaction (3)) with a yield of 6%. It produces formic acid at a rate of 0.1% h^{-1} for ethylene under the condition of 100 ppb ozone. The CH_2O_2 radical reacts with HCHO and H_2O (reaction (4)) to produce formic acid.

Another pathway is the oxidation of formaldehyde by HO_2 and OH radicals, respectively. The former reaction is estimated to be 10^{-3}% h^{-1} for the rate of disappearance of HCHO, smaller than reaction (3) in the polluted air, and the contribution of the reaction with OH to be much less. Thus, the mono-carboxylic acids were presumed to be produced by the oxidation of olefins by ozone.

For the formation of di-carboxylic acids, cyclic olefins and aliphatic diolefins are supposed to be the important precursors (Grosjean, 1977; Grosjean et al., 1978; Grosjean and Friedlander, 1980; Hatakeyama et al., 1985 and 1987). They are written as



$$(CH_2)_{n-2}\text{(CH=CH)} + O_3 \rightarrow C_{n-1}\text{ diacid}$$

$$+ OH \rightarrow C_n \text{ diacid}$$

$$CH_2{=}CH(CH_2)_{n-4}CH{=}CH_2 + O_3 \rightarrow C_{n-2}\text{ diacid}$$

For the production of succinic acid photooxidation of cyclopentene is considered to be an important pathway. However, about 70 ppt of cyclopentene precursor is required, for the succinic acid concentration to reach as high as 100 ng m^{-3} in the daytime (Hatakeyama et al., 1987). Oxidation of aliphatic diolefins may be also important for di-carboxylic acid formation. However, for the kinematic analysis, neither reaction rate constants nor reaction pathways have been clear yet and further investigation are waited for.

5. Conclusion

The behavior of mono- and di-carboxylic acids during the long-range transport of photochemical air pollution from the metropolitan, large emission source area along the Tokyo Bay to the inland mountainous region in central Japan was investigated as a part of a cooperative field observation of meteorology and chemistry in summers of 1983 and 1986. Mono- and di-carboxylic acids were measured every three hours at inland observation sites along the transport route of the polluted air mass.

Of the mono-carboxylic acids except for formic acid, acetic acid was the most abundant species, comprising 90% of the total. The concentrations of acetic acid, propionic acid and *n*-butyric acid increased to the ranges of 4–8 ppb, 0.3–0.7 ppb and 0.1–0.3 ppb, respectively, in the daytime. In particular, the concentrations attained maximum levels as the polluted air mass arrived at the observation sites, while they decreased at night. These diurnal variations were similar to that of ozone.

Of the di-carboxylic acids succinic acid was the most abundant species in the airborne aerosol, followed by phthalic acid and malonic acid. The concentrations ranged from 20–130 ng m^{-3}. Diurnal variations were also similar to that of ozone. The concentrations of di-carboxylic acids were much higher in the daytime than at night. This suggests that most of the mono-carboxylic acids and almost all of the di-carboxylic acids were produced by the photooxidations of anthropogenic compounds during long-range transport.

5. Acknowledgements

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