

## DETERMINATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN THE ICE CORE SAMPLES FROM GREENLAND

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**Abstract:** Polycyclic aromatic hydrocarbons (PAHs) were determined in the ice core samples taken from Greenland, Site-J (205 m long, ca. 450 years old) using a combined gas chromatography and mass spectrometry (GC-MS). More than forty PAHs ranging from naphthalene to benzo(ghi)perylene were detected in the ice samples. Phenanthrene, methylphenanthrene and dimethylphenanthrene were among major PAH species. Concentrations of total PAHs were low (av., 58 ng/kg-ice) before 1920 whereas they started to increase after the 1920s (av., 970 ng/kg-ice for the 1920s to 1980s) and gave the highest value (5800 ng/kg-ice) in the late 20th century. These trend is primarily explained in terms of an increased anthropogenic emission to the atmosphere from fossil fuel combustion in the northern hemisphere as well as atmospheric transport of the combustion-derived products over the Greenland ice sheet. This study demonstrated that PAHs are useful tracers to study atmospheric transport of anthropogenic pollutants and to reconstruct the historical trend of global air pollution.

### 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous organic contaminants in the environment, which are emitted to the atmosphere by fossil fuel combustion and biomass burning (e.g., SIMONEIT and MAZUREK, 1981; GRAEDEL *et al.*, 1986). The PAHs have been studied in atmospheric samples such as rain (MAZUREK and SIMONEIT, 1986; KAWAMURA and KAPLAN, 1986a), snow (KAWAMURA and KAPLAN, 1986b), and aerosols (SICRE *et al.*, 1987; SIMONEIT *et al.*, 1991) and been successfully used as chemical tracers of anthropogenic input to the atmospheric environment. They have been detected in aquatic environments such as lacustrine (WAKEHAM *et al.*, 1980) and marine sediments (HITES *et al.*, 1977; LIPIATOU and SALIOT, 1991). Combustion-derived PAHs have also been studied in Arctic aerosol particles (DAISEY *et al.*, 1981). Gaseous and particulate PAHs originated from civilized areas in mid-latitudes are transported to the Arctic regions through the atmosphere and are scavenged by wet and dry precipitation. Thus, the transported PAHs should be preserved in glaciers and ice sheets. However, the PAH compounds have only rarely been studied in ice core samples (GREGOR, 1991), although ionic species and gases have been extensively studied recently (e.g., DELMAS, 1992).

In this paper, we studied a 205 m long ice core collected from Greenland for the distribution of PAH species using gas chromatography–mass spectrometry. The PAHs were used as chemical tracers of fossil fuel combustion and biomass burning to reconstruct historical changes of the human influence on the Arctic atmospheric environment for the last 450 years.

## 2. Samples and Analytical Procedures

The ice core samples (205 m long) were taken from Site-J, Greenland (66°51.9'N, 46°15.9'W, altitude: 2030 m) in 1989 with a mechanical shallow drill by the Japanese Arctic Glaciological Expedition 1989 (WATANABE and FUJII, 1990). The ice layer chronology was determined by electrical conductivity and volcanic eruption records: the 205 m layer was estimated to be *ca.* 450 yr old (SHOJI *et al.*, 1991). The ages of the surface layers were confirmed by the determination of tritium (IZUMI, a personal communication, 1991). Twenty six sections collected from the core samples were used in this study for the PAH analyses. The length of each section was 0.2–1.5 m, which corresponds to less than one half year to 4 years. Most of the ice sections were less than the average annual accumulation rate (42 cm of ice or 38 cm of water, SHOJI *et al.*, 1991). To avoid potential contamination, the sample surfaces were removed from each ice section and then melted in a pre-cleaned Teflon container in the clean room. The melt water was poisoned with mercuric chloride to avoid microbial degradation of organic compounds. The samples were stored at 4°C in a pre-cleaned brown glass bottle with a Teflon-lined screw cap prior to analysis, which prevents gas exchange with the ambient air. A procedural blank was prepared using pure water.

A melt water sample (*ca.* 350 ml) was taken in a 500 ml separatory funnel and acidified with 5 ml 6 M HCl, which was pre-extracted with methylene chloride to remove organic contaminants. Organic compounds were extracted with a methylene chloride/ethyl acetate (2:1) mixture (70+40+40 ml). Extracts were combined, concentrated and then saponified with a 0.5 M KOH/methanol solution (10 ml) containing 20% water in a glass ampule at 100°C for 60 min. The methanol in the saponified fraction was removed by using a rotary evaporator under a vacuum and then pure water was added. Neutral compounds were isolated from the acidic components in the saponified fraction by solvent extraction with n-hexane/methylene chloride (2:1). The neutrals were concentrated and subjected to silica gel column chromatography. PAHs were eluted with an n-hexane/methylene chloride mixture (2:1). The PAH fraction was concentrated, transferred into a 1.5 ml screw vial with a Teflon-lined cap, dried with a nitrogen stream and dissolved in n-hexane (30  $\mu$ l).

The organic solvents used were distilled in the laboratory using all-glass apparatus prior to use. Pure water was prepared by distillation after oxidizing organic impurities of Milli-Q water with potassium permanganate. Silica gel (Bio-Sil A, 200-400 mesh, Bio-Rad Labs.) was extracted with methanol and methylene chloride, dried under vacuum using a rotary evaporator (water-bath temperature gradually increased from 20 to 80°C) and activated in an oven at

210°C for 24 hours. The silica gel was deactivated with 1% pure water. After occasional shaking for 2 days, the gel was stored in hexane. The silica gel column was prepared by packing the gel in a Pasteur pipet (4 cm high) with quartz wool.

For the determination of PAHs, a computerized GC-MS system (Finnigan-MAT ITS 40) was used, to which a fused silica capillary column (DB-5, 0.25 mm i.d. × 30 m long, film thickness: 0.25 μm) was installed. The column temperature was programmed from 50°C (2 min) to 120°C at 30°C/min and then to 310°C (20 min) at 6°C/min. Mass spectra were obtained every one second in a mass range of  $m/z$  50–600 and the data were processed with a COMPAQ work station (Model DESKPRO 386/20e). Identification of PAHs was performed by comparison of mass spectra and retention times with authentic standards (26 species) as well as mass spectral information stored in the computer (NIST library) and published in the literature (SIMONEIT *et al.*, 1991). Quantification was performed by comparing the peak areas of the mass chromatogram at  $m/z=M^+$  (molecular ions) with those of authentic PAHs which were used as external standards. The recoveries of 26 PAHs (from naphthalene to benzo(ghi)perylene) were 77–99% (av. 93%) in the analytical procedures including a solvent extraction to GC-MS measurement. Duplicate analyses showed that the analytical error was within 7%. The concentrations reported here are corrected for the procedural blanks, which include the processes from melting of the ice sample to GC-MS determination.

### 3. Results and Discussion

#### 3.1. Distribution of PAHs in the Greenland ice core

Figure 1 shows a reconstructed ion chromatogram of the PAH fraction isolated from the ice core sample (depth 6.7–7.1 m, age: 1980). More than forty PAHs ranging from naphthalene ( $mw=128$ ) to 2,9-dimethylpicene ( $mw=306$ ) were identified in the ice samples. Major PAH species detected include biphenyl, fluorene, methylfluorene, dimethylfluorene, dibenzothiophene, methyl-dibenzothiophene, phenanthrene, methylphenanthrene, and dimethylphenanthrene. The molecular distributions of PAHs in the samples, especially after 1900s A.D. showed an abundant presence of alkyl-substituted structures such as methyl, dimethyl and trimethyl substituted PAHs in addition to parent compounds. Interestingly, the alkyl substituted PAHs contain positional isomers of the alkyl group as seen in Fig. 2, which gives examples of mass chromatograms at molecular ions ( $m/z=M^+$ ) of phenanthrene and its alkylated derivatives. The alkylated derivatives and their positional isomers were generally detected for the parent PAH compounds such as naphthalene, fluorene, dibenzothiophene and phenanthrene in the ice samples. Anthracene, benzo(b)fluorene, benz(a)anthracene, benzo(k)fluoranthene, benzo(e)pyrene and benzo(ghi)perylene were also detected in the samples as relatively minor components, although they are not listed in Table 1.

The PAH constituents detected in the ice samples are present in crude oils and have been reported in the combustion products of fossil fuels (*e.g.*, GRAEDEL *et al.*, 1986). The alkyl-substituted PAHs have also been abundantly reported in

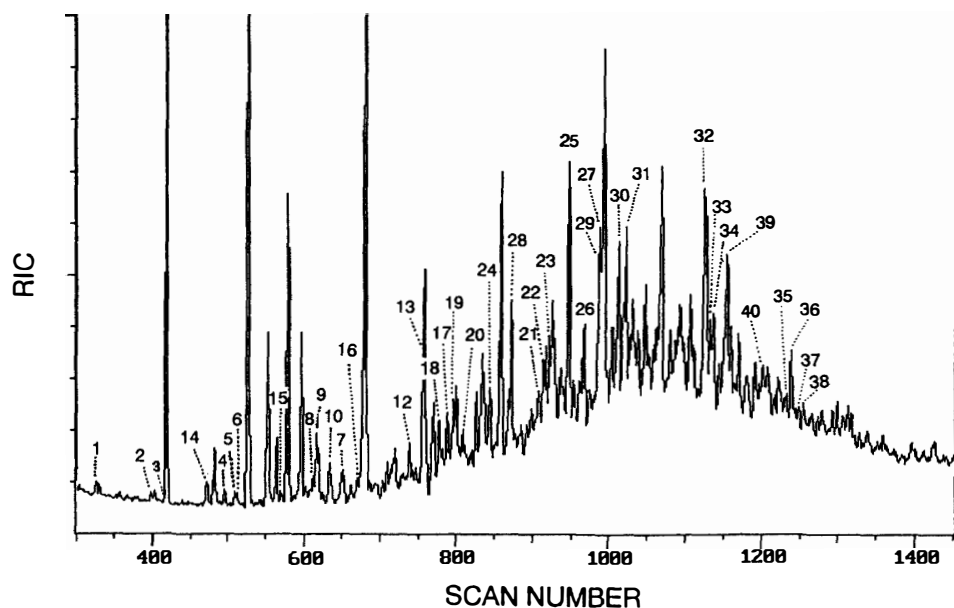


Fig. 1. Reconstructed ion chromatogram of polynuclear aromatic hydrocarbon fraction extracted from the ice core sample collected from Greenland (#10, 6.7–7.1 m in depth, age: 1980). For the compound names, see Table 1.

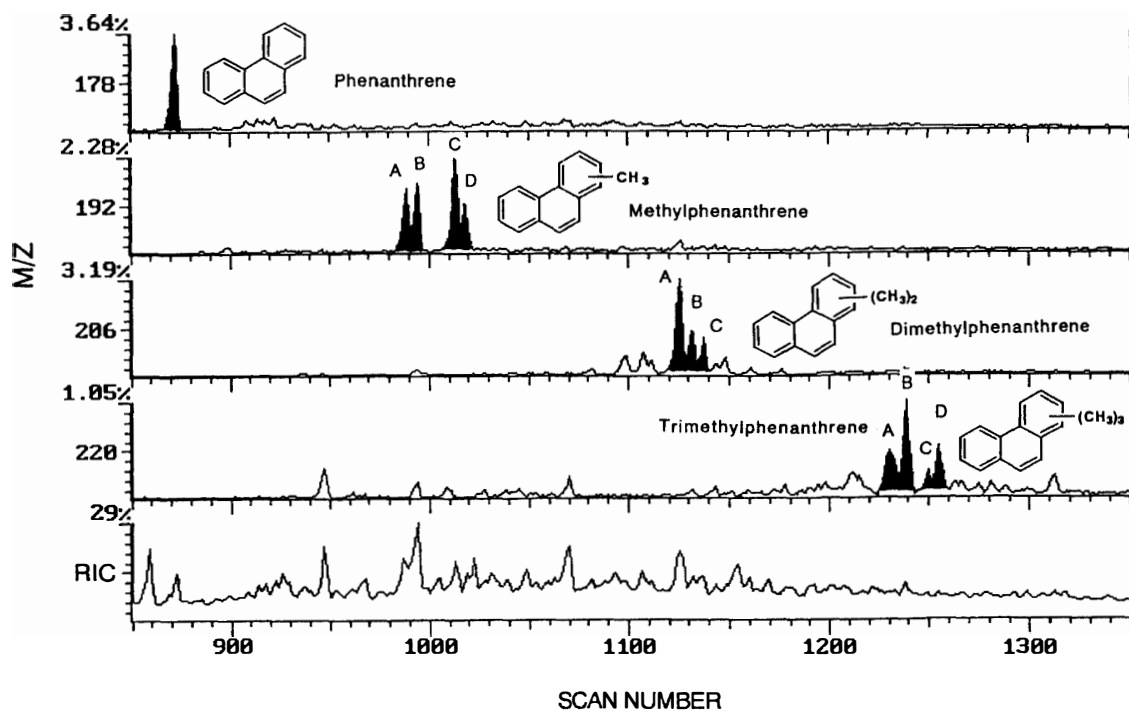


Fig. 2. Mass chromatograms of phenanthrene and alkyl-substituted phenanthrenes separated from the ice core sample collected from Greenland (#10, 6.7–7.1 m in depth, age: 1980).

exhausts from internal combustion engines (JENSEN and HITES, 1983; LEE and SCHUETZLE, 1983). Similar distributions of the PAH constituents detected in ice samples have been reported in the polluted atmospheric gas and/or particulate phases (YAMASAKI *et al.*, 1982). Thus, the PAHs in the ice core samples are

Table 1. Polynuclear aromatic hydrocarbons (PAHs) detected in the Greenland ice core (Site-J) and their concentration ranges and mean concentrations.

No	Name of PAH	M.W.	Concentrations (ng/kg-ice)			
			Minimum	Maximum	Mean	Median
1	Naphthalene	128	0	9.8	1.4	0.22
2	2-Methylnaphthalene	142	0	12	2.5	0.68
3	1-Methylnaphthalene	142	0	11	1.7	0.39
4	2,6-Dimethylnaphthalene	156	0	13	2.1	1.0
5	Dimethylnaphthalene A	156	0	18	2.7	1.3
6	Dimethylnaphthalene B	156	0	10	1.9	1.1
7	2,3,5-Trimethylnaphthalene	170	0	57	6.8	1.5
8	Trimethylnaphthalene A	170	0	29	3.1	0.73
9	Trimethylnaphthalene B	170	0	41	3.4	0.02
10	Trimethylnaphthalene C	170	0	52	6.1	1.5
11	Trimethylnaphthalene D	170	0	25	3.0	0.78
12	Tetramethylnaphthalene A	184	0	63	6.7	1.2
13	Tetramethylnaphthalene B	184	0	60	6.2	0.59
14	Biphenyl	154	0	21	3.6	1.9
15	Acenaphthene	154	0	10	1.3	0.39
16	Fluorene	166	0	36	4.3	1.9
17	2-Methylfluorene	180	0	110	9.9	1.4
18	Methylfluorene A	180	0	79	7.8	0.81
19	Methylfluorene B	180	0	114	11	1.8
20	Methylfluorene C	180	0	31	2.9	
21	Dimethylfluorene A	194	0	204	20	1.9
22	Dimethylfluorene B	194	0	156	18	2.4
23	Dimethylfluorene C	194	0	194	21	1.4
24	Dibenzothiophene	184	0	121	13	1.4
25	Methyldibenzothiophene A	198	0	773	74	6.6
26	Methyldibenzothiophene B	198	0	635	58	5.1
27	Methyldibenzothiophene C	198	0	458	42	4.2
28	Phenanthrene	178	0	275	33	8.6
29	Methylphenanthrene A	192	0	230	23	2.7
30	Methylphenanthrene C	192	0	361	38	5.6
31	Methylphenanthrene D	192	0	179	18	2.4
32	Dimethylphenanthrene A	206	0	525	47	5.5
33	Dimethylphenanthrene B	206	0	216	21	2.6
34	Dimethylphenanthrene C	206	0	175	16	2.1
35	Trimethylphenanthrene A	220	0	132	9.2	
36	Trimethylphenanthrene B	220	0	172	15	1
37	Trimethylphenanthrene C	220	0	37	3.5	
38	Trimethylphenanthrene D	220	0.38	87	7.6	0.45
39	Fluoranthene	202	0	23	2.2	0.62
40	Pyrene	202	0	24	3.4	0.8
41	Chrysene/Triphenylene	228	0	0.65	0.05	
42	2,9-Dimethylpicene	306	0	28	8.0	0.9
	Total		1.5	5850	584	94

M.W.=Molecular weight. A, B, C, and D mean positional isomers which were not specified due to a lack of authentic standards. "0" in the minimum column means below the detection limit (*ca.* 0.1 ng/kg-ice).

primarily derived from anthropogenic sources. The organic pollutants emitted to the atmosphere in mid-latitudes of the northern Hemisphere are subjected to long-range transport over Arctic regions, especially in winter (BARRIE and BOTTENHEIM, 1991). The PAHs transported over the Arctic are likely scavenged from the air by snow flakes and dry deposition, and preserved in the Greenland ice sheet.

The PAH compositions in the ice core samples are characterized by predominance of relatively low molecular weight PAHs (such as phenanthrene,  $mw=178$ ). This is in contrast to the molecular distributions of PAHs in aerosol samples collected from mid-latitude areas, which usually indicate the predominance of higher molecular weight PAHs such as fluoranthene ( $mw=202$ ), pyrene ( $mw=202$ ) and benzo(a)pyrene ( $mw=252$ ) (SIMONEIT and MAZUREK, 1981). The PAH distributions in wet precipitation samples, however, have shown abundant presence of lower molecular weight species such as naphthalene ( $mw=128$ ) and its methyl derivatives (KAWAMURA and KAPLAN, 1986a). Further, lower aromatic hydrocarbons are important species in the gas phase of the ambient atmosphere (YAMASAKI *et al.*, 1982). These considerations suggest that the PAHs detected in the Greenland ice core samples are scavenged from the Arctic atmosphere to the ice sheet largely by wet (snow) precipitation, which removes PAHs present in both gaseous and particulate forms. Alternatively, dry deposition may also be important during the winter season when temperature is low enough to cause the gaseous hydrocarbons to be adsorbed on the aerosol particles.

### 3.2. Changes in PAH distributions in the last 400 years

The concentration ranges for the PAH species in 26 samples from the Greenland ice core are summarized in Table 1, as well as mean and median concentrations. Total concentrations of the PAHs detected in the ice core sample ranged from 1.5 ng/kg-ice to 5850 ng/kg-ice. The lowest concentration was observed in the ice layer of 181.8 m in depth, which corresponds to 1602 A.D. By contrast, the highest value was obtained at the depth of 6.7–7.1 m, which corresponds to 1980 A.D. Figure 3 presents total PAH concentrations as a function of age in the ice core layers. The PAH concentrations are very low ( $30\pm 22$  ng/kg) in the 16th to 18th centuries, slightly high ( $98\pm 100$  ng/kg) in the 19th century and very high ( $910\pm 1600$  ng/kg) in the 20th century. Although they scattered with time, the concentrations seem to increase significantly from pre-industrial to industrial ages. These results suggest that more aromatic hydrocarbons were emitted to the atmosphere after the 1920s, probably by enhanced fossil fuel combustion in the Northern Hemisphere, and were transported to south Greenland.

However, low PAH concentrations were sometimes observed, even in the 1940s to 1980s (Fig. 3). Such low values do not indicate decreased fossil fuel combustion; rather they suggest that polluted air masses were less abundantly transported over the south Greenland ice sheet. One explanation is that a change in meteorological conditions such as atmospheric circulation patterns in a certain period may potentially have occurred and depressed the long-range transport of

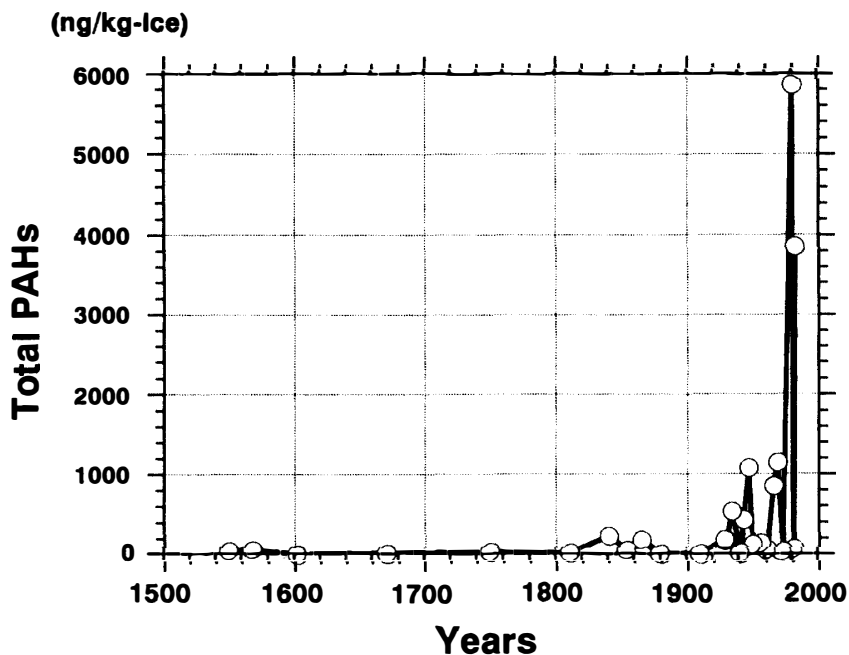


Fig. 3. Concentrations of total PAHs in the Greenland ice core (Site-J) as a function of age.

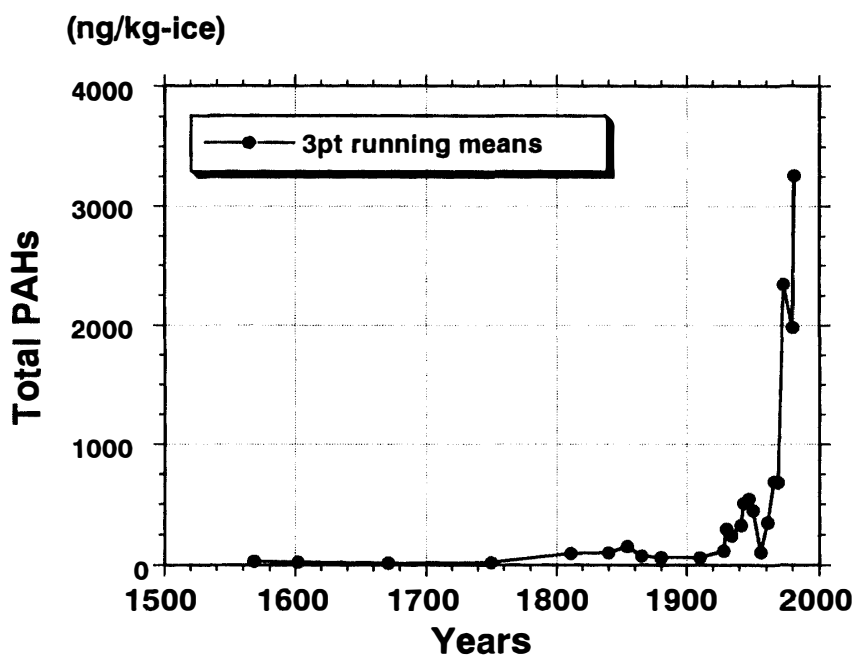


Fig. 4. Historical trend of total PAH concentrations in the Greenland ice core (Site-J, Greenland). The concentrations are presented as three point running means.

the polluted air mass over south Greenland. Alternatively, these fluctuations may partly be involved with the lengths of ice core cuts. Because the ice cut length is sometimes less than 20 cm, which is far less than the averaged annual accumulation rate (42 cm of ice), some samples cover less than half a year and do not

contain snow layers accumulated in the winter season. In the Arctic, the atmospheric transport of polluted air from mid-latitude is enhanced during winter to early spring based on observations of sulfate and nitrate (BARRIE and BOTTENHEIM, 1991). Thus, the snow layers accumulated in the winter should contain more organic pollutants including PAHs, and vice versa.

To minimize the fluctuations in the PAH concentrations probably caused by the short length of the ice section and the seasonality of the polluted air mass transport as stated above, running means of the concentration data were taken. Figure 4 presents the change in the PAH concentrations in the ice core as a function of age on the basis of running three point means. The results clearly show a historical trend of PAH concentrations in the ice core, that is, the concentrations were very low before the 1900s, started to increase drastically around the 1910s and reached the highest concentrations in the 1980s. This trend demonstrates that the atmospheric transport of anthropogenic organic matter was enhanced after the 1930s mainly due to the enhanced emission of fossil fuel combustion products to the atmosphere. Relatively high concentrations are observed in the 1800s, in which naphthalene and its alkyl substituted species as well as biphenyl increased preferentially. These PAHs in the ice core may have a source different from other constituents such as phenanthrene and its alkylated derivatives, which did not show a peak in the 1800s.

The general features of the PAH concentrations in the ice core are in good agreement with the historical record of world petroleum production, as shown in Fig. 5. World crude production increased exponentially from 1900 A.D. to 1980 A.D., suggesting enhanced oil combustion and subsequent emission of organic pollutants to the atmosphere. The present study clearly demonstrates that the combustion-derived pollutants were transported to the Arctic regions and preserved in ice layers, providing informations on historical fossil fuel usage. Enhanced activity of human beings has been recorded in the acidity of the same ice core samples (FUJII and KAMIYAMA, unpublished data, 1991): the pH decreased from 5.3 (1910 A.D.) to 5.15 (1980 A.D.). The measurements of non-sea salt sulfate and nitrate in the south Greenland ice core (Dye 3) also indicated increased concentration of these inorganic pollutants in the last 100 years (MAYEWSKI *et al.*, 1986).

Although the general trends in the concentrations of PAHs are consistent with petroleum production rates, a drastic decrease in their concentrations was observed around the 1950s to 1960s (Fig. 4). This apparent discrepancy is probably caused by past changes in meteorological conditions, with reduced atmospheric transport of polluted air to south Greenland. At this moment, we cannot explain how the atmospheric circulation patterns have changed over Greenland and other Arctic areas. However, it is interesting to note that such a decrease was also observed in the concentrations of biogenic fatty acids (KAWAMURA *et al.*, unpublished results) and (non-sea salt) excess sulfate (FUJII and KAMIYAMA, unpublished data, 1991). These results suggest that the distributions of organic compounds preserved in the Greenland ice core also reflect a change in meteorological conditions which is probably involved with a change of the air



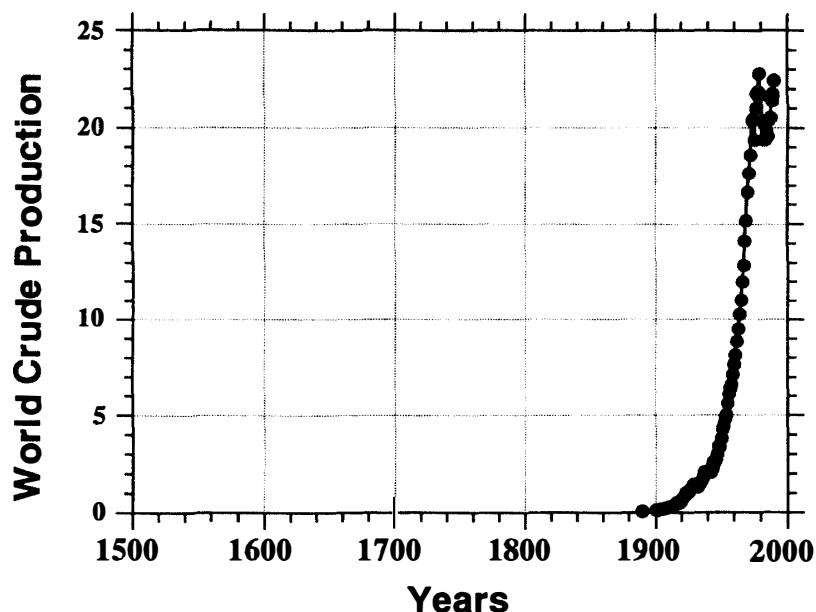


Fig. 5. Historical change in the world crude production. The plotted data were taken from TWENTIETH CENTURY PETROLEUM STATISTICS (1991).

circulation pattern in high latitude on either a local or global scale.

#### 4. Concluding Remarks

More than forty polynuclear aromatic hydrocarbons (PAHs) have been detected in a Greenland ice core (Site-J, *ca.* 400 years old) by using a GC-MS. Major PAHs detected were phenanthrene and methyl-, dimethyl- and trimethyl-substituted phenanthrenes. Alkylated PAHs were generally as abundant as the parent compounds. Their molecular distributions in the ice core indicated that PAHs were derived from fossil fuel combustion and carried to south Greenland by atmospheric transport. The concentrations of total PAHs were low in 1550–1800 A.D. They increased slightly in 1800–1900 A.D. In contrast, the PAH concentrations started to increase considerably in 1920s and showed the highest value in the 1980s. These changes are generally consistent with world crude production records. However, lower values of PAH concentrations were observed in the 1950s in spite of the continuous increase in fossil fuel production and usage in the world. This suggests that atmospheric transport of pollutant organic matter over the Greenland ice sheet has largely been reduced by a drastic change in the atmospheric circulation pattern over the Arctic polar region. The present study showed that the PAHs preserved in an ice core are excellent tracers to reconstruct the past changes of fossil fuel usage and long range atmospheric transport over the Arctic region.

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