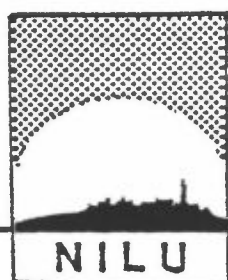


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ORGANIC AIR POLLUTANTS IN THE NORWEGIAN ARTIC

by

Jozef M. Pacyna
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ROYAL NORWEGIAN COUNCIL FOR SCIENTIFIC AND INDUSTRIAL RESEARCH

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P.O. BOX 130, N-2001 LILLESTRØM
NORWAY

ISBN 82-7247-685-1

SUMMARY

Atmospheric emission sources and transport within air masses of some polychlorinated hydrocarbons, fluoranthene and quinones are reviewed and their importance for the Arctic is discussed. The origin of hydrocarbons in the Arctic is examined with the help of measured concentration ratios of γ -HCH to α -HCH (HCH - hexachlorocyclohexane) during 5 measurement campaigns between Fall 1982 and Summer 1984 at stations in the Arctic and the Norwegian mainland, and information from weather maps and wind back-trajectories. Emissions from sources in the Soviet Union contributed significantly to the high episodic concentrations of measured hydrocarbons in the Arctic both during wintertime and summertime. Emission sources in Europe influenced the concentrations at the Norwegian mainland stations during summertime. Pollutants from these and North American sources can, however, also be traced in the Norwegian Arctic. Local sources contributed mainly to the concentrations of fluoranthene and quinones.

Deposition of hydrocarbons "en route" and in the Arctic is also discussed as well as the γ - to α -HCH isomerization.

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ORGANIC AIR POLLUTANTS IN THE NORWEGIAN ARCTIC

1 INTRODUCTION

In recent years, there has been a growing interest in studying the transport of organic compounds to the Arctic. The work of Rasmussen and Khalil (Khalil and Rasmussen, 1983; Rasmussen et al, 1983) has shown that a number of organic gases which are transported into the Arctic originate from combustion processes in the more densely populated and in industrialized regions. These gases may contribute significantly to the haze formation. Oehme and Ottar (1984) indicated that organic air pollutants are brought into the Arctic together with other pollutants and deposited by precipitation and dry deposition. In summer the air concentrations of the more volatile compounds seem to be significantly influenced by sublimation of previously deposited materials.

The content of various organic compounds can be used to trace the origin of polluted air masses. In order to confirm this, samples of gaseous and particulate organic matter were collected at various stations in the Norwegian Arctic and mainland Norway in the period 1982-1984. The concentrations of α - and γ -hexachlorocyclohexane (α -HCH and γ -HCH), hexachlorobenzene (HCB), pentachlorobiphenyl (5 Cl-biphenyl), α -chlordane, fluoranthene and quinones (phenanthroquinones and anthraquinones) are presented here and their possible origins are discussed. From the summer 1983 measurement campaign, concentrations of pentachlorobenzene (PeCB) are also available.

It was considered an advantage to select organic polychlorinated compounds which show large differences in their physical and chemical characteristics, such as chemical stability, vapour pressure, water solubility, biodegradation rate, volatilization from soil or water and atmospheric residence time. The choice was made on the basis of information given by Lyman et al. (1981). A further requirement was the simultaneous analysis of all compounds by a single gas-chromatographic/massspectrometric determination to minimize the costs of the analysis.

Since the behaviour of organic compounds in the air and their emission sources are not as well documented in the literature as for inorganic gases and particulate matter, a short review of emission and atmospheric transport of chlorinated hydrocarbons, fluoranthene and quinones is given in the following chapter of this report.

2 SOURCES AND ATMOSPHERIC TRANSPORT OF CHLORINATED HYDROCARBONS

2.1 EMISSION SOURCES RELATED TO THE USE OF PESTICIDES

The main source of chlorinated hydrocarbons in the air is the application of pesticides to increase food and forest production. There are several ways for pesticides to enter the atmosphere.

2.1.1 Spray application

The major injection of pesticides into the air takes place during the spraying of agricultural crops. The pesticide spray is disseminated into the atmosphere in the form of finely divided droplets or particles, of optimum size for maximum control of insects or weeds. This is not always achieved and the spray usually consists of droplets or particles with a wide range of sizes. In addition, the liquid carriers may evaporate under certain conditions, changing the particle size.

The parameters affecting the quantity of pesticide that reaches a target include particle size distribution, amount of material sprayed, physical-chemical properties of the pesticide as well as height at which the material is released, wind speed, and atmospheric turbulence.

Cunningham (1962) has reported that more than 40% of the original aqueous drop is lost by evaporation before the droplets have fallen 11 m, a very important problem, especially in summer heat. Sometimes evaporation suppressants are added to water-pesticide mixtures to diminish the vaporization losses.

2.1.2 Volatilization from crops and soils

Evaporation of pesticide compounds may continue after treating agricultural soils and plant foliage. This adds to the above described releases during spray application and is believed to account for almost 90% of the pesticide contamination in the air (Lewis and Lee, 1978). For example, DDT is lost from soil mainly due to evaporation, and the loss is most intensive immediately after application.

There are several factors influencing the rate of loss by volatilization. The losses are faster from wet than dry soils, and from sandy than high clay soils. Water increases the vapour pressure of the pesticide by competing for adsorption sites. Thus, the mass flow of water moving to the surface of the soil during evaporation contributes to the greater pesticide evaporation from wet than dry soils. Other factors include temperature and humidity of the air, and wind velocity. Old pesticides, deposited during the previous growing seasons, may also be released into the atmosphere when a field is plowed for the next crop.

Pesticides evaporate also from plant surfaces. Pesticide sprays are deposited predominantly on the foliage, and the total surface area of the foliage is often many times that of the soil surface. The pesticides do not penetrate deeply into the leaf surface, and as a result they are exposed to volatilization, dislocation by rain, and photooxidation. One interesting difference of pesticide evaporation from crops and soils is that increased air humidity slightly decreases the former process and increases the latter one.

2.1.3 Evaporation from water

Surface waters, such as rivers, lakes and oceans, are the main sinks for the majority of air pollutants, among them the pesticides. After deposition, much of the pesticidal material is bound to sediments and only a small part is in a true solution.

The evaporative loss of pesticides from water surfaces is limited to compounds with a moderate vapour pressure and a low solubility. The most commonly studied pesticide in this respect is DDT. Peak use and production of DDT occurred in the U.S. in the late 1950's and early 1960's (Woodwell et al., 1971). Use was banned in the U.S. and Canada in 1972, however, until 1982, the U.S. exported DDT to the Middle and Far East, Africa and Latin America (Rapaport et al., 1985). Generally, the major input of several pesticides into the ocean is via the atmosphere and not the rivers (Lewis and Lee, 1976).

2.1.4 Manufacturing processes

The production of pesticides is also a likely source of pesticide emissions to the atmosphere. Production of the various particles involves the use of many flammable solvents, and occasional fires have been reported (Lewis and Lee, 1976) causing a threat to nearby residents due to pesticide penetration into residences.

2.1.5 Statistical information on the production and use of pesticides considered in this report

There is rather limited information on the use of various pesticides containing chlorinated hydrocarbons in the literature. Lindane (purified γ -HCH isomer) is widely used in various parts of the world (Mathys, 1984 - personal communication), however, statistical information on its application is scarce. FAO statistics (1979) shows that huge amount of HCH have been used by the countries during the last decade. India used ca. 7.7×10^4 t technical HCH during 1975-1977 (Tanabe et al., 1982). Technical HCH consists roughly of 70% α -HCH, 7% β -HCH, 13% γ -HCH, 5% σ -HCH and 5% of other impurities (Tatsukawa et al., 1972; Baumann et al., 1980). According to Jakes and Vemuri (1980), pesticides, including HCH, are likely to be used on an increasing scale in India in the future to improve the agricultural crop yield and for the eradication of diseases. Wang (1981) informs about the production and use of HCH in China over a long period. An amount of 2×10^4 t of technical HCH is produced annually in the Shenyang Chemical Plant alone (Anonymous, 1981). Japan used 4.10^5 t of technical HCH from 1948 to 1970. Although, accurate data on the world production and use of HCH are not yet

available, it is safe to assume that the major consumption areas of HCH are concentrated in the northern hemisphere, especially on the Asian Continent. The above presented numbers for India, China and Japan should be considered together with the consumption of HCH in the Soviet Union. Komarov (1980) indicated that it is huge, but no statistics are available.

Olafson (1978) reported a large consumption of lindane in eastern Australia where it has been applied in the sugar cane growing regions adjacent to the Great Barrier Reef.

Lindane is also used in Europe. In Poland, lindane is used as a fungicide (the mixture includes among others 20% pure γ -HCH and 2.5% of Hg) and to control many soil pesticides, as reported by Janicki (1976). He also suggested that γ -HCH is used in Poland to keep parking lots, airports, roads and railways clean of insects. Janicki (1976) indicates that 2.9×10^3 t of γ -HCH is used for these purposes in the United Kingdom. This amount seems to be overestimated when comparing with the agriculture application of γ -HCH. In Norway, 6.6 t of lindane was used in 1982 and the γ -HCH content was not less than 99% according to Paulsen (1984, personal information). About 9 t of lindane was used in Denmark in 1981 (Kemikaliekontrollen, 1982) and 6 t in Sweden in the same year (Bekämpningsmedel, 1982). In Finland, lindane is used as an agriculture insecticide and a forest pesticide (Tiittanen and Blomquist, 1982). The first application consumed 7.3 t lindane, with only 49 kg of the active ingredients, in 1981. Of three forest pesticides in use, one is applied for the protection of unbarked timber against insect infestations and the two others are used to protect planted pine seedlings against the large pine weevil, *Hylobius abietis*. All three contain lindane as the active ingredient. The amount used in 1981 was 24.6 t pesticides containing 4.9 t lindane and these values have not changed significantly from year to year. No information exists on this lindane application in other countries.

In the EPPO (European and Mediterranean Plant Protection Organization) area lindane is widely used for disinfection of empty storage rooms (Mathys, 1984), however, no numbers are available.

2.2 EMISSION SOURCES OF FLUORANTHENE AND QUINONES

The two main sources of fluoranthene are transportation and fuel combustion in stationary sources (Brown, 1981). The U.S. EPA (1978) has estimated emission factors of fluoranthene to 66.8 $\mu\text{g}/\text{l}$ for diesel vehicles and 230 $\mu\text{g}/\text{l}$ for gasoline-powered cars.

Coal combustion contributes to the emissions of fluoranthene and emission factors in the range 90 to 270 $\mu\text{g}/\text{t}$ coal as can be suggested on the basis of data by Wilson et al. (1980). Considering statistical information on the world consumption of gasoline, diesel oil and coal, including lignite (UN, 1985), and the above emission factors, the fluoranthene emissions from transportation are significantly larger than emissions from coal combustion in stationary sources, on a global scale. Thus, fluoranthene seem to be a good tracer for emissions from transportation.

Both antraquinones and phenanthroquinones can be released during combustion processes (incineration, power generators), but they are also present in soils (quinones are decomposition products of biological materials). These compounds are very useful in the detection of a possible influence of local sources on measurements at background stations.

2.3 PHYSICAL FORM OF CHLORINATED HYDROCARBONS IN THE ATMOSPHERE

The physical form of the pesticides in the atmosphere depends upon their residence time in the atmosphere, chemical and physical properties (e.g., surface tension, vapour pressure and photoreactivity), environmental factors (e.g. temperature, humidity, wind turbulence) and on the nature and concentration of airborne particles.

There is very little published information on the physical form of pesticides in the atmosphere. This lack of data is largely due to a bias introduced by the sampling techniques. Many of the early measurements indicated that chlorinated hydrocarbons as pesticide residues in the atmosphere existed predominantly in the form of suspended particulate matter (e.g., Tabor 1965; Antommara et al., 1965). Later, with the progress made in the sampling techniques, it was found, that several pesticides are present in

significant amounts in the vapour state (e.g., Bjorklund et al., 1970). Beyermann and Erkrich (1974) indicated that the mean quantity of gaseous lindane in the air over Mainz, West Germany was twice as large as the dust-associated part. As the substantial vaporization loss of pesticides from soil and vegetation has already been documented, likely most of the airborne residues of lindane are in the vapour phase. A part of these gaseous molecules is absorbed onto suspended particles and the efficiency of the sorption process is dependent on the frequency of collisions between particles and gaseous molecules and the surface properties of the particle. If the particle is wet, the adsorption will be determined by the water solubility of the pesticide, which is low. Since the concentrations of chlorinated hydrocarbons are present in ng.m^{-3} or pg.m^{-3} and the concentrations of suspended particles in $\mu\text{g.m}^{-3}$, the adsorption becomes low. As suggested by Lewis and Lee (1976), highly carbonaceous material, such as that emitted from coal-burning stacks adsorbs more polychlorinated hydrocarbons than silicate particles, such as airborne sandy soil. Thus, during the transport of anthropogenic pollutants a part of the gaseous pesticides will be adsorbed on the particles, particularly those with diameter lower than $2.0 \mu\text{m}$, but for the more volatile pesticides most of the material may be in the gas phase.

2.4 LONG-RANGE TRANSPORT OF CHLORINATED HYDROCARBONS WITHIN THE AIR MASSES

There is much good evidence in the literature on the long-range transport of chlorinated hydrocarbons within the air masses, starting with the work by Cohen and Pinkerton in 1966 on the translocation of pesticides by air transport and rain-out. Global transport via the atmosphere was considered to be the only explanation for the presence of pesticides in Antarctica (Peterle, 1968) Barbados (Risenbrough et al., 1968; Seba and Prospero, 1971; Bidleman et al., 1981) and in the air over the North Atlantic as far as 2000 km from the U.S. coast (Harvey and Steinhauer, 1974). DDT and HCH found in the North Pacific have been transported from the Asian continent by the westerly winds, as concluded by Tanabe and Tatsukawa (1980).

Several mechanisms of long-range transport of chlorinated hydrocarbons have been postulated (e.g., Pooler, 1971; Woodwell et al., 1971). The primary transport mechanism suggested is turbulent diffusion which depends on wind vectors, temperature and topography. Gaseous compounds also undergo the simultaneous process of molecular diffusion, but this is much less significant than turbulent diffusion under most meteorological conditions (Lewis and Lee, 1976). Pooler (1971) studied global circulation problems and found that they play a major role in the long-range transport of airborne pesticide residues. His simplified schematic diagram of the hemispheric circulation pattern is shown in Figure 1. The mean meridional circulation is the dominant feature in the subtropics, between equator and 30° latitude. The equatorward flow is present at low altitudes in the tropical easterlies, and a compensating poleward flow aloft. As indicated in Figure 1, the subsidence in the subtropical high pressure zones at ca. 30° latitude completes the circulation. The level flows from the northern and southern hemispheres meet in the intertropical convergence zone, where the principal exchange of air masses takes place. Poleward of this intertropical zone, the eddy transport by migratory high and low pressure systems dominates the circulation at higher altitudes. Pooler (1971) indicated that the mean feature of flow at higher latitudes is the eastward motion of the air at all levels, except high in the stratosphere. Thus, the parcel of air may circulate for a long time before it leaves the latitude of origin, and compounds with a long residence time in the atmosphere may be transported over very long distances. This applies to many of the chlorinated hydrocarbons. For example, Atkins and Eggleton (1970) estimated the residence time of lindane to be 17 weeks. Atlas and Giam (1981) calculated the residence time of PCB to be 189 days.

2.5 REMOVAL PROCESSES

Chlorinated hydrocarbons are removed from the air by various physical and chemical processes. The former group of processes includes removal by precipitation and dry deposition. Atkins and Eggleton (1970) studied the removal of lindane, dieldrin and DDT from the atmosphere and found a weekly removal of lindane of 2.5 per cent by precipitation and 3.3 per cent by dry deposition. The wet removal of dieldrin and DDT was 0.1 and 0.4 per cent, respectively, and dry deposition accounted for 3.5 and 3.4 per cent per week.

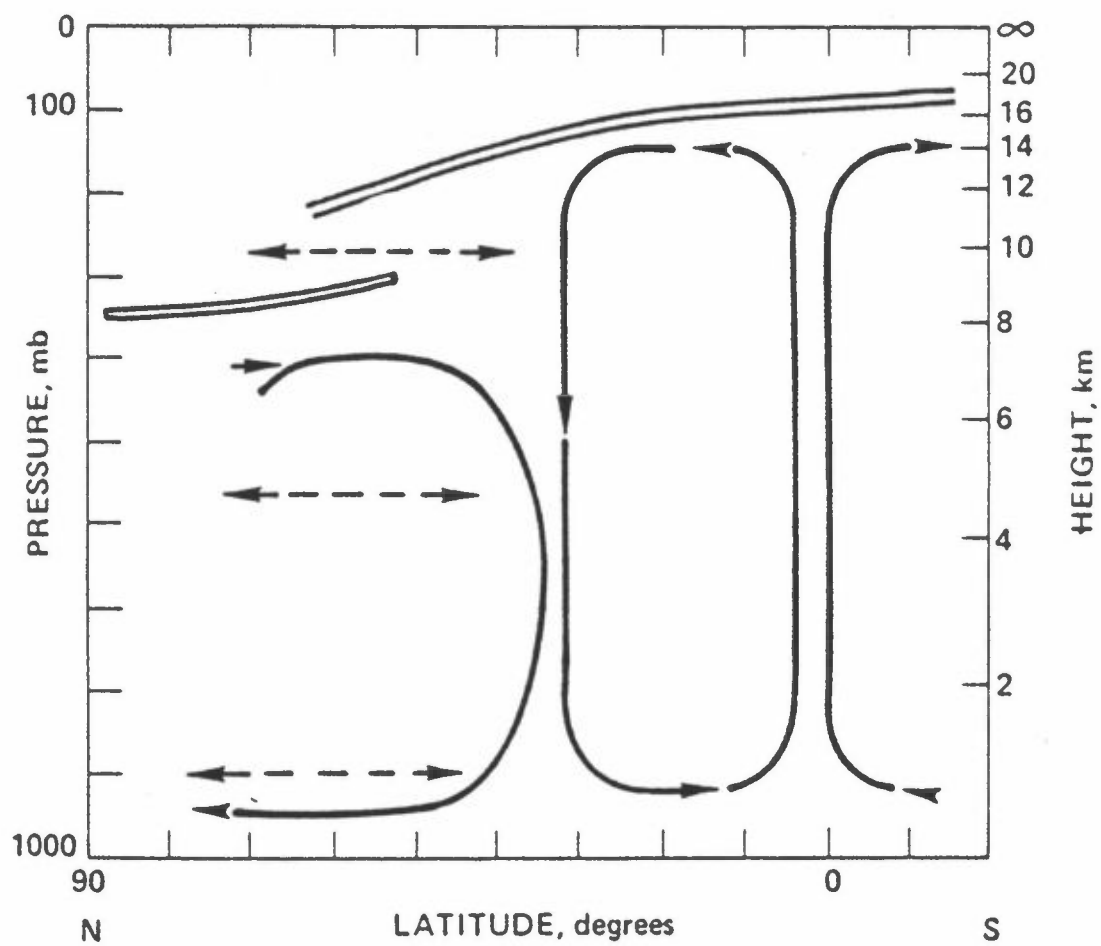


Figure 1: Hemispheric meridional circulation pattern (adapted from Pooler (1971)).

The deposition velocities of individual chlorinated hydrocarbons differ by 1-2 orders of magnitude, even at the same sampling site (Bidleman and Christensen, 1979). Mc Clure (1976) observed an increase of the dry deposition velocity (V_d) of PCBs from 0.3 to 3.0 cm.s^{-1} with wind speed for particles $>1.0 \mu\text{m}$ in diameter. For submicron particles ($<1.0 \mu\text{m}$ i dia.) this effect is not important (e.g., Sehmel and Sutter, 1974). About 20-30% of the polycyclic aromatic hydrocarbons in the atmosphere is concentrated on the $>1.0 \mu\text{m}$ dia. particles (Bidleman and Christensen, 1979). Because of the much higher deposition velocities of large particles (Sehmel and Sutter, 1974) the flux of high-molecular weight organics might be expected to be large-particle dominated and hence greatly influenced by wind speed. Other important parameters in this respect include ambient temperature and changes in the atmospheric particulate load. Bidleman and Christensen (1979) concluded that higher ambient temperatures would be expected to reduce the adsorption of organics to aerosols and to increase their re-volatilization. Generally, higher values of V_d were found for the less volatile organic compounds, reflecting a greater degree of attachment to aerosols.

The wet deposition process is often characterized by washout ratios ($\mu\text{g.m}^{-3}$ in rain per $\mu\text{g.m}^{-3}$ in air). For polychlorinated hydrocarbons these ratios are generally low (1.5×10^3 for HBC, $1-5 \times 10^4$ for HCH). However, Bidleman and Christensen (1979) found that measured washout ratios were significantly higher than those calculated from the equilibrium partitioning of the organic compounds between air and water. They suggested that while PCB and DDT in the atmosphere are partitioned between the vapour and particle phases, they are removed during precipitation by washout or rainout of particles and not by vapour dissolution in raindrops. Similar conclusions for PCB wet removal were forwarded by Murphy and Rzeszutko (1977) and Slinn et al. (1978).

Among a few chemical processes leading to the removal of chlorinated hydrocarbons from the atmosphere, photodegradation is the only mechanism that may compete with physical removal processes. The hydroxy radical plays a major role in the photodegradation of chlorinated hydrocarbons in the atmosphere. It can react with the various pesticides to form peroxides, epoxides and ketones. Ozone, singlet state oxygen and atomic oxygen also react with various organic compounds.

So far, most of the investigations have been performed in the condensed or adsorbed phases of organic compounds, beginning with the photodegradation studies of synthetic pesticide by Wichmann et al. (1946) who discussed the decomposition and volatility of DDT. Later the information on the vapour-phase photolysis of pesticides appeared in the literature (e.g., Nagl et al., 1970; Crosby and Moilanen, 1974). Sometimes the products of photodegradation of pesticides in the air are less desirable than the parent compounds. For example, photodieldrin is more toxic than dieldrin. Many organochlorine pesticides resist photolysis as they resist metabolism (Lewis and Lee, 1976). Others lose their chlorine only slowly. One of the most photosensitive pesticides is chlordane (Baker and Applegate, 1974). Lindane is slightly less photosensitive than chlordane.

3 EXPERIMENTAL

3.1 SAMPLING PROCEDURE

Glass fiber filters of 142 mm diameter (Gelman, Type 61635, Ann Arbor, MI., USA) were precleaned at 400⁰C in a muffle furnace for 3 h and further by Soxhlet extraction in hexane for 8 h. Polyurethane foam [foamed from technical grade toluenediisocyanate and a polyoxypropylenetriol (Caradol 42-2, Shell Company) after standard procedure] was supplied by a local factory. A 5 cm thick sheet with a density of 25 kg.m⁻³ was cut into plugs of 11 cm diameter. The plugs were afterwards repeatedly squeezed in toluene at 100⁰C, and then extracted for 12 h, first with pesticide grade acetone and then with hexane. After drying them at 70⁰C under vacuum in a desiccator for 48 h the plugs were stored in tightly wrapped aluminium foil. Previously used plugs can be prepared for use again after an extraction in acetone and hexane for 8 h.

The sampler (see Figure 2) was thoroughly cleaned with pesticide grade acetone and hexane before use. The sampling speed was in the order of 24 m³.h⁻¹. After sampling, both the glass fiber filter and the foam plugs were wrapped in aluminium foil and sent to the laboratory. When no immediate analysis was possible, the samples were stored at -20⁰C.

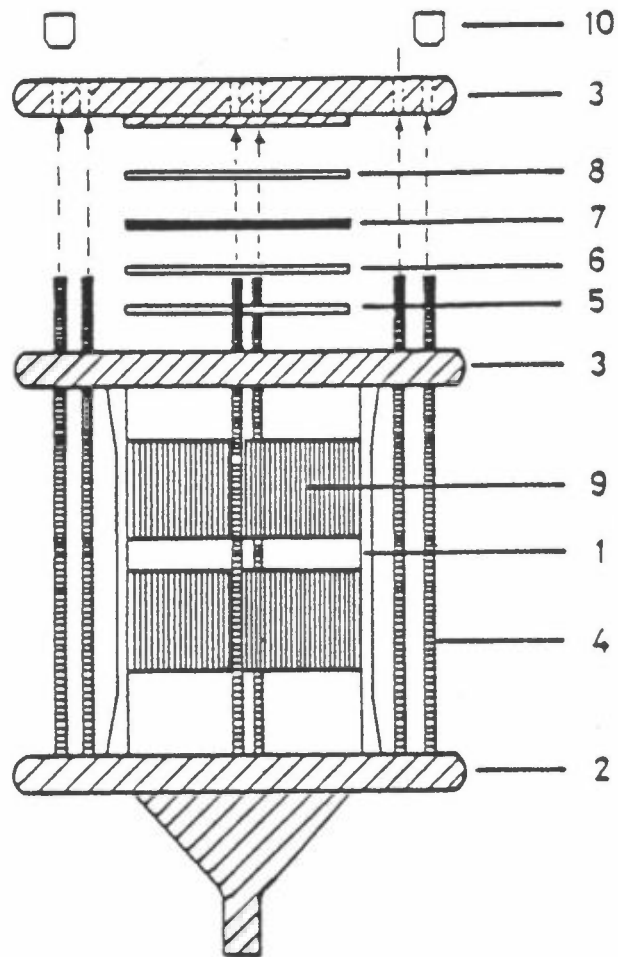


Figure 2: Construction of the high volume PUR sampler. 1 Glass tube with flat buttress ends, 20 cm long, 10 cm i.d. (Pipeline Component PS 4/200 Corning Limited, England); 2 Aluminium outlet adapter for the high volume pump; 3 aluminium flange; 4 screw bolt; 5 silicon washer; 6 aluminium screen (filter support); 7 glass fiber filter; 8 Teflon gasket; 9 foam plugs; 10 set nut.

3.2 CHEMICAL ANALYSES

Sample Pretreatment and Clean-up

The glass fiber filter and the PUR foam plug were Soxhlet-extracted with 100 and 300 ml of hexane +10% diethylether, respectively, for 8 h. The extract was concentrated to about 200 μ l and the whole sample was fractionated on a highly active silica by HPLC. Activation of the column was done by pumping a mixture of hexane/acetic acid/2,2-dimethoxypropane (90+10+2.5) through the column. One main fraction was collected using hexane +0.4% acetonitrile as solvent. It contained alkanes, polycyclic aromatic hydrocarbons and the chlorinated compounds of interest. The elution time of endrin was used as a cut-off criteria for the collection.

Gas-Chromatographic Separation

Capillary Columns. Glass capillary columns made of soda lime glass with an internal diameter of 0.3 mm and a length of 30 m, were used. The glass surfaces were leached with HCl and deactivated by persilylation. The columns were statically coated with OV1, (film thickness of about 0.15 μ m) and helium was used as carrier gas. Separation numbers (C_{12} - C_{13}) were around 30-35.

Separation Conditions. Injection: 1 μ l splitless or on-column injection. Temperature program: injection at 40⁰C, 1 min at 40⁰C, 40-80⁰C with 30⁰C.min⁻¹, 80-250⁰C with 5⁰C.min⁻¹ and 250⁰C isothermal.

Quantification

The mass spectrometric analysis was carried out on a 5985B GC/MS system equipped with a 5840 gas chromatograph (Hewlett-Packard, Palo Alto, Calif., USA), and a home-made GC/MS interface with a fused silica capillary as transfer line. A home-made reagent gas inlet was used for negative ion chemical ionization.

Quantification was carried out using negative ion chemical ionization with methane as reagent gas at 0.4 torr and 200⁰C. The multiple ion detection

mode was used with a dwell time of 50 ms for each ion. The detection limit (signal-to-noise ratio 10:1) was in the order of 1 pg for most compounds.

Further information about the method is given in Oehme and Stray (1982) and Oehme (1982).

Samples were collected at four arctic island stations and five Norwegian mainland stations. The station location is presented in Figure 3.

4 MEASUREMENT RESULTS

The result of the hydrocarbon measurements made under the BP project are presented in the form of time series plots for all the intensive measurement campaigns at the different ground stations in Figures 4 through 26. These are 2- and 3-day mean concentrations of α - and γ -HCH, HCB, 5-PCB, α -chlor-dane, fluoranthene and quinones (sum of phenanthroquinones and anthraquinones).

All Hi-Vol sampling runs were conducted on a 2+2+3 day schedule, with the first 2-day run normally starting on a Monday morning, and the last 3-day run ending on Monday morning.

For quick reference, all intensive campaign periods are listed in Table 1.



Figure 3: Locations of BP Project ground stations.

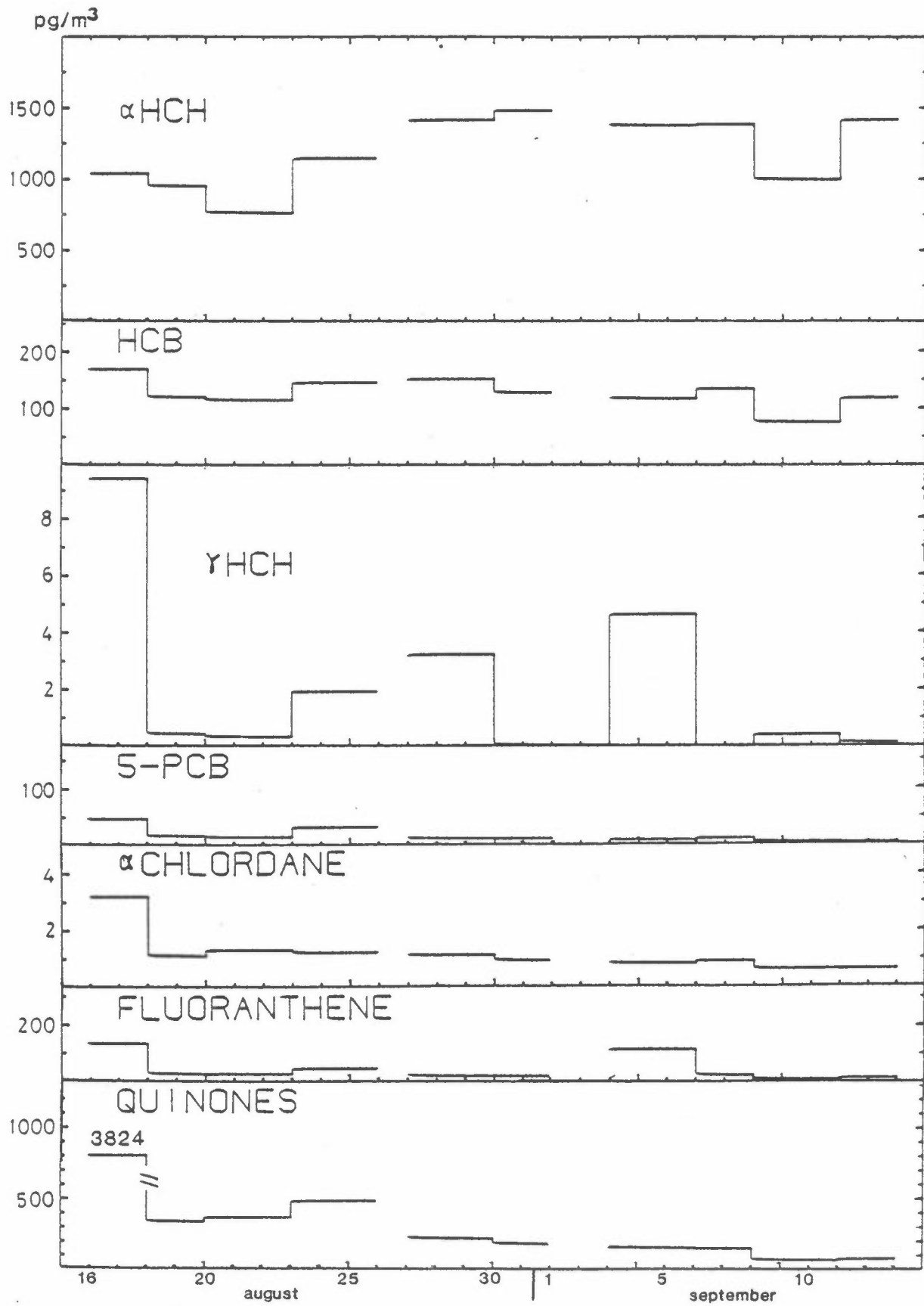


Figure 4: Time series of 2- and 3- day mean concentrations of hydrocarbons at Ny Alesund (NYA) during the Fall 1982 campaign.

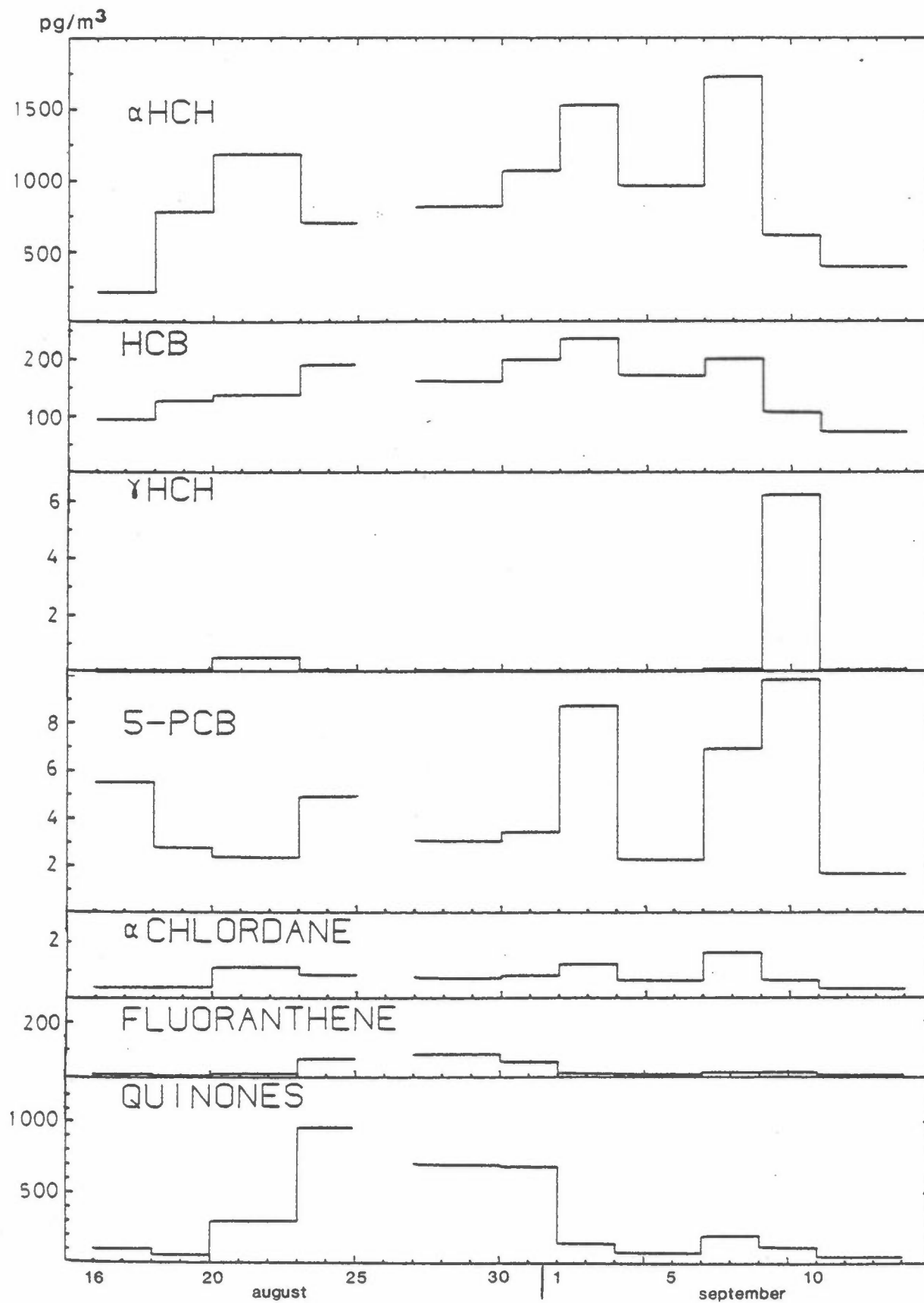


Figure 5: Time series of 2- and 3- day mean concentrations of hydrocarbons at Hopen (HOP) during the Fall 1982 campaign.

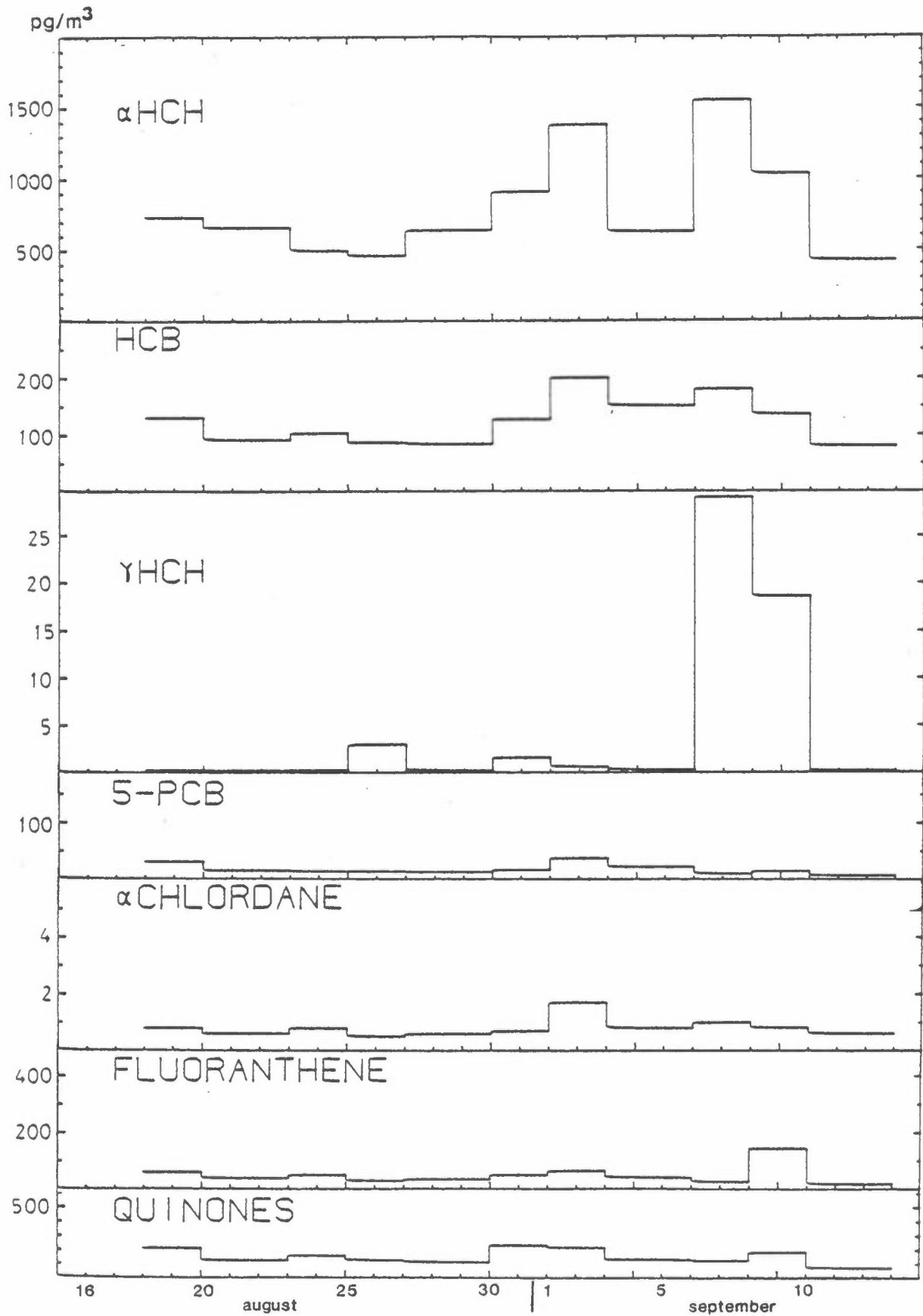


Figure 6: Time series of 2- and 3- day mean concentrations of hydrocarbons at Bjørnøya (BJO) during the Fall 1982 campaign.

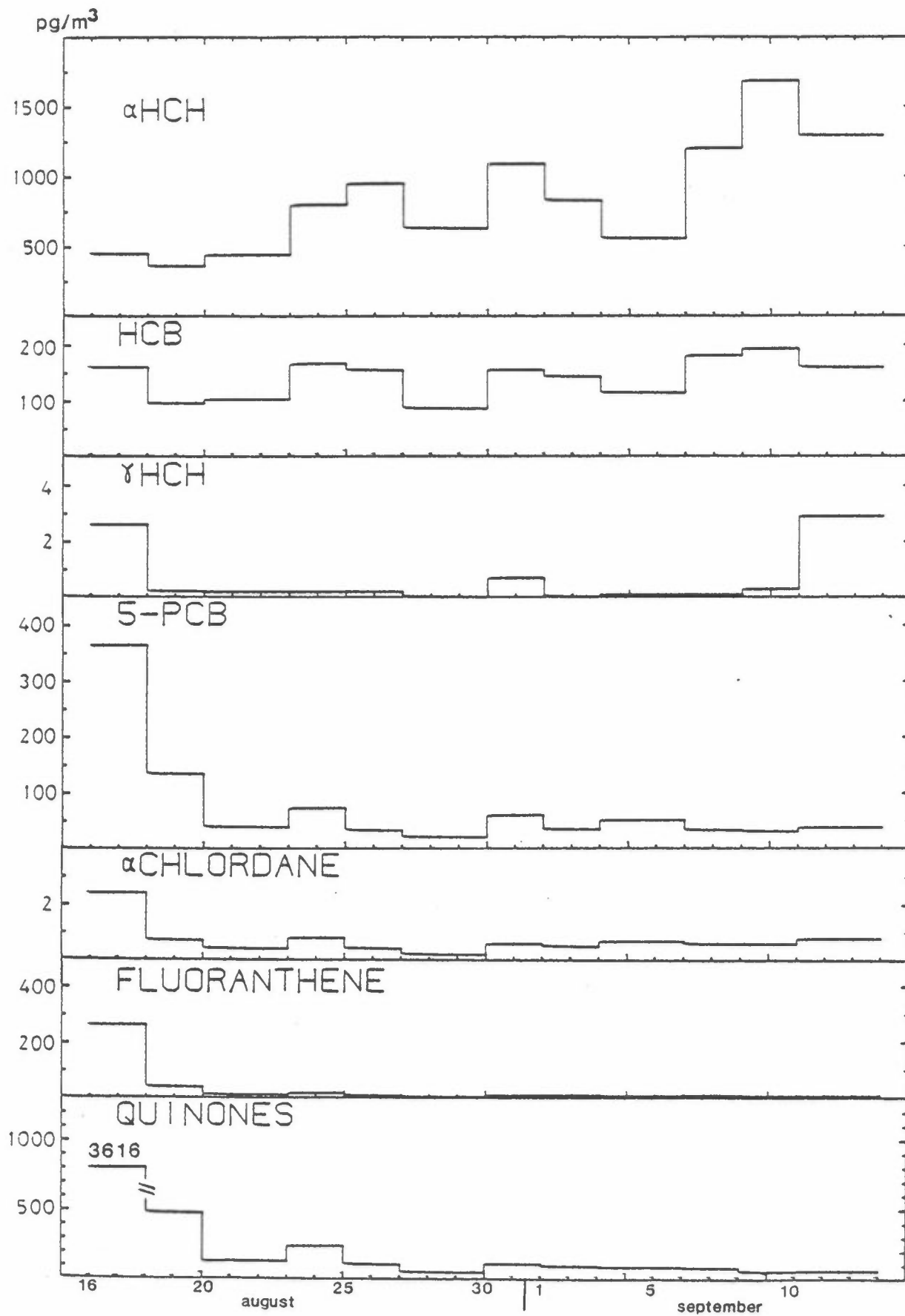


Figure 7: Time series of 2- and 3- day mean concentrations of hydrocarbons at Jan Mayen (JAM) during the Fall 1982 campaign.

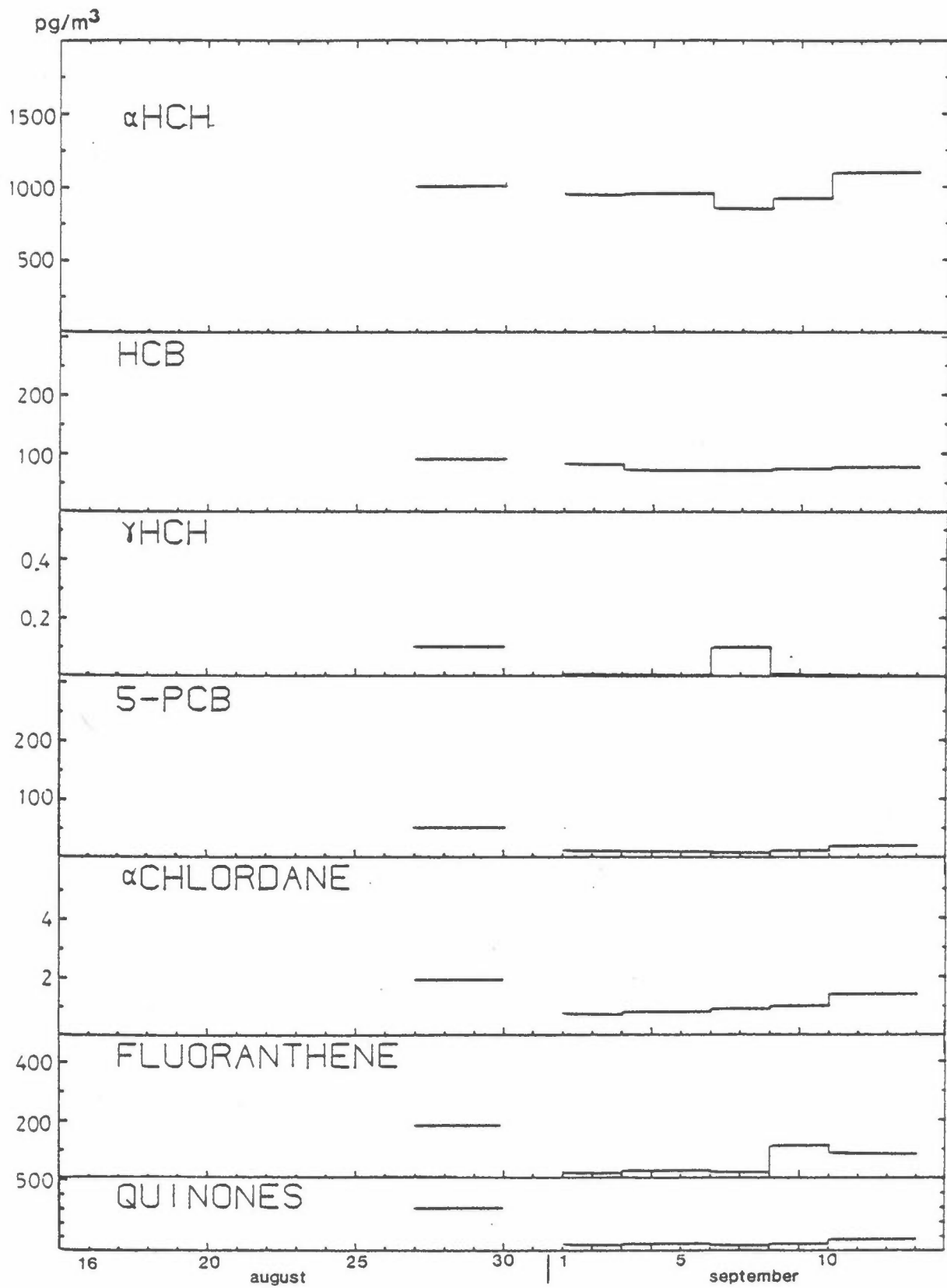


Figure 8: Time series of 2- and 3- day mean concentrations of hydrocarbons at Birkenes (BIR) during the Fall 1982 campaign.

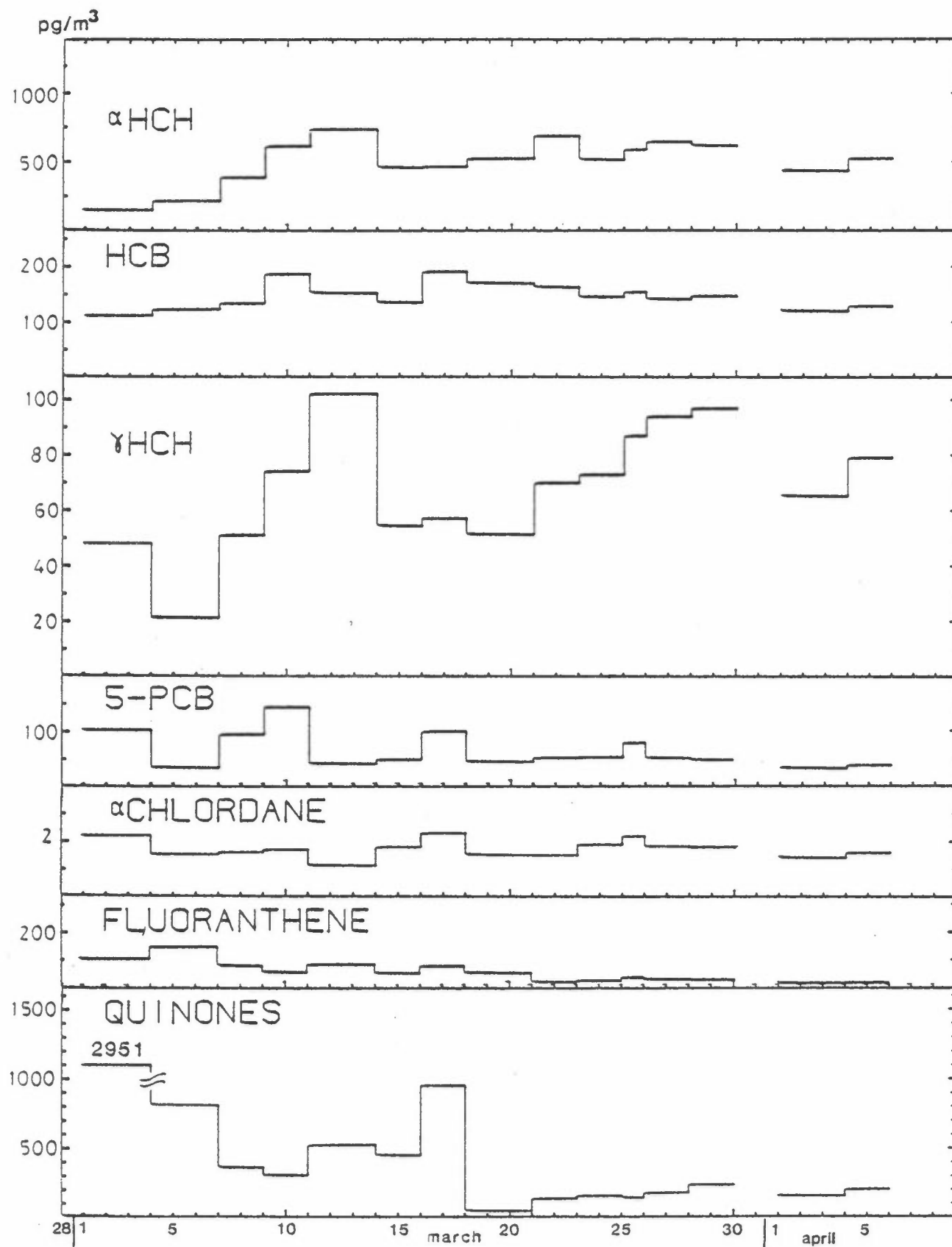


Figure 9: Time series of 2- and 3- day mean concentrations of hydrocarbons at Ny Alesund (NYA) during the Winter/Spring 1983 campaign.

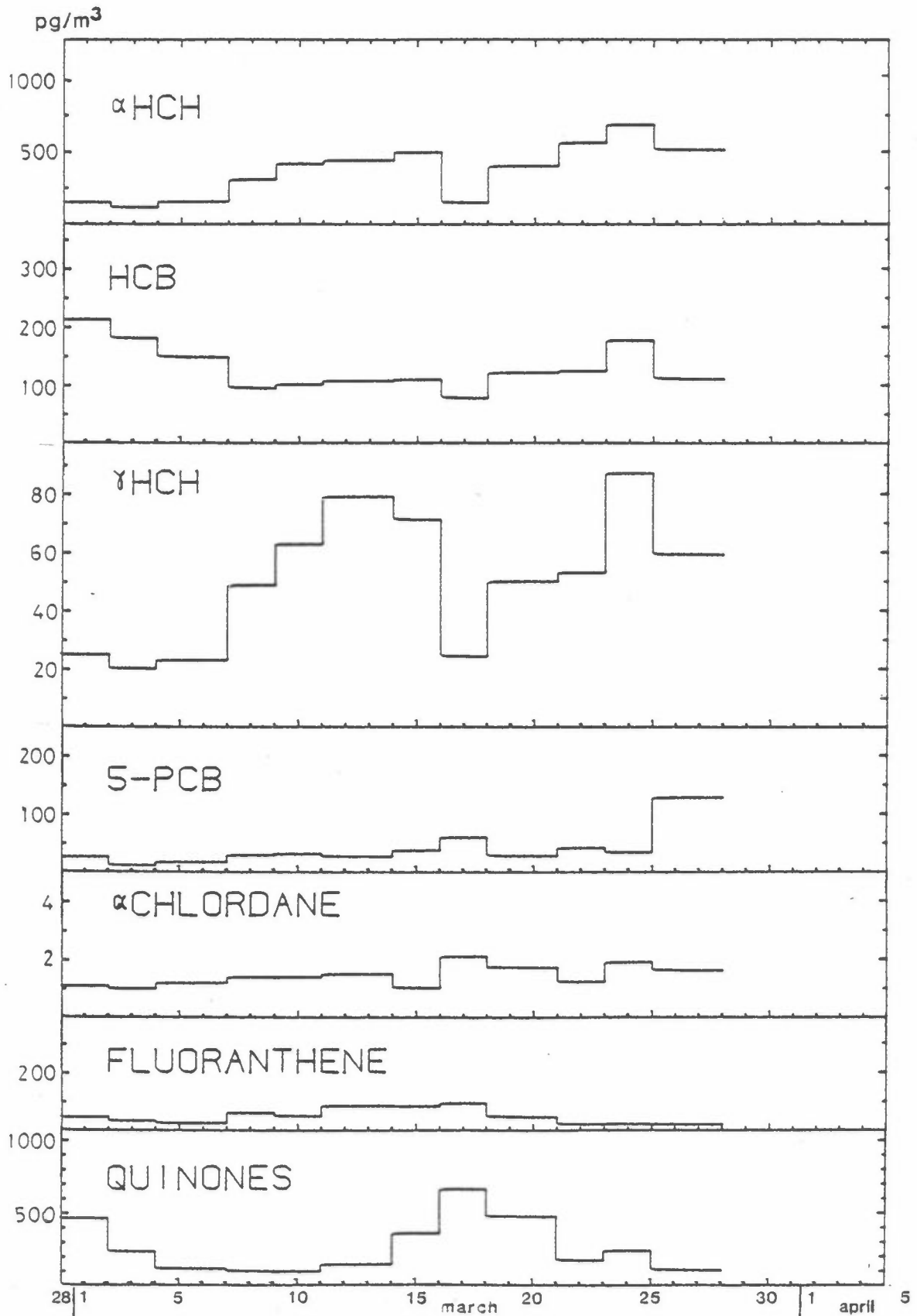


Figure 10: Time series of 2- and 3- day mean concentrations of hydrocarbons at Hopen (HOP) during the Winter/Spring 1983 campaign.

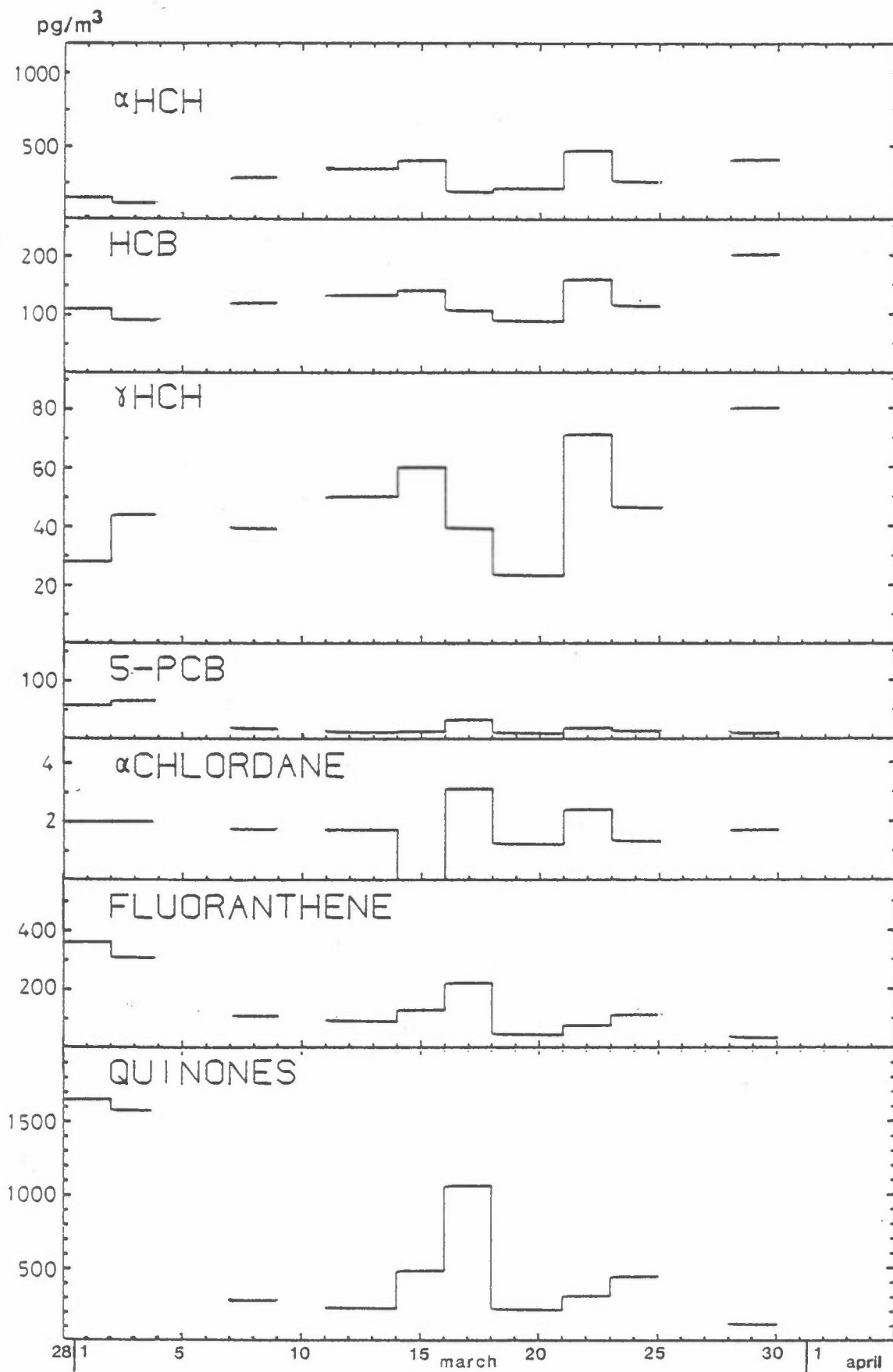


Figure 11: Time series of 2- and 3- day mean concentrations of hydrocarbons at Bjørnøya (BJO) during the Winter/Spring 1983 campaign.

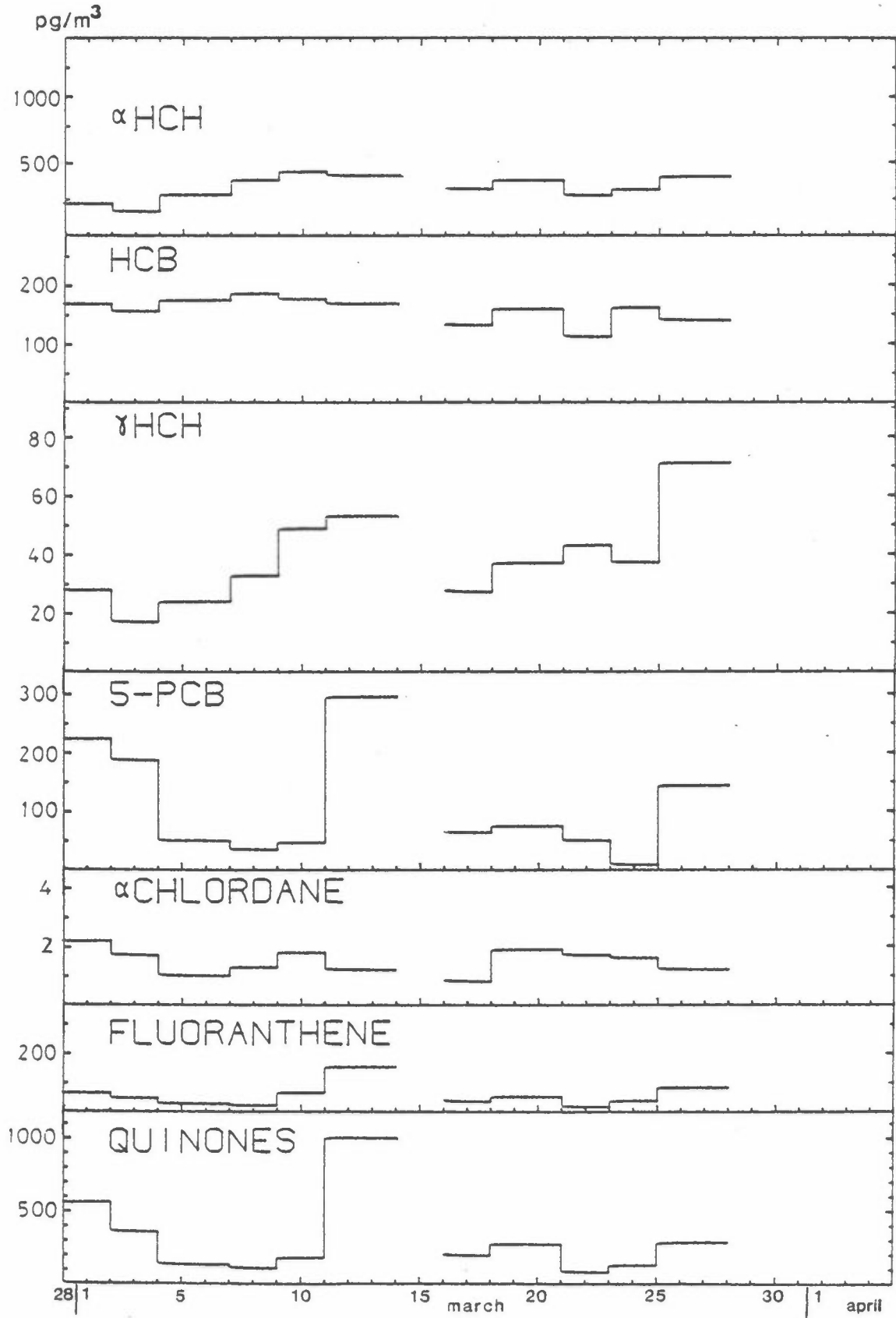


Figure 12: Time series of 2- and 3- day mean concentrations of hydrocarbons at Jan Mayen (JAM) during the Winter/Spring 1983 campaign.

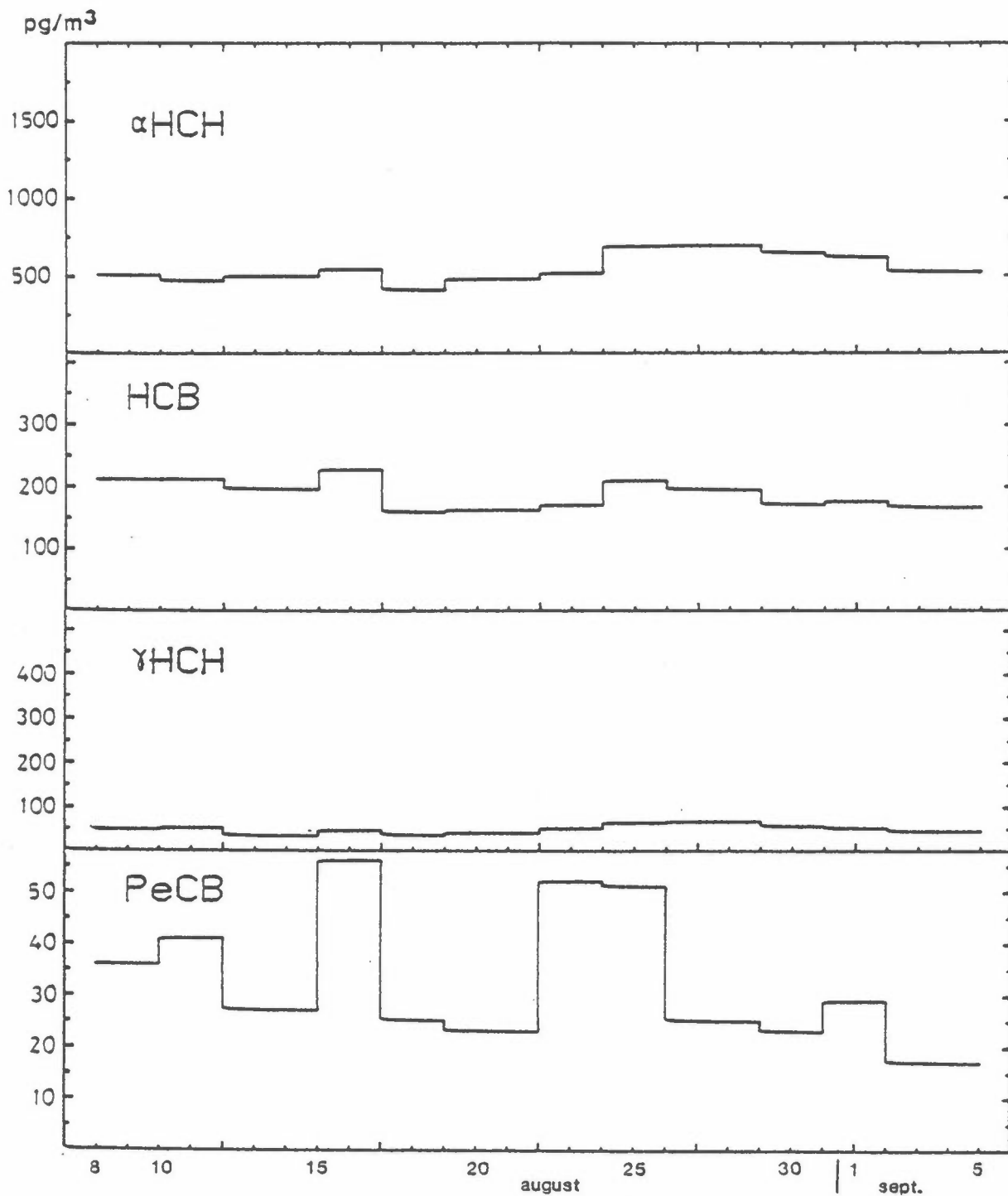


Figure 13: Time series of 2- and 3- day mean concentrations of hydrocarbons at Ny Alsesund during the Fall 1983 campaign.

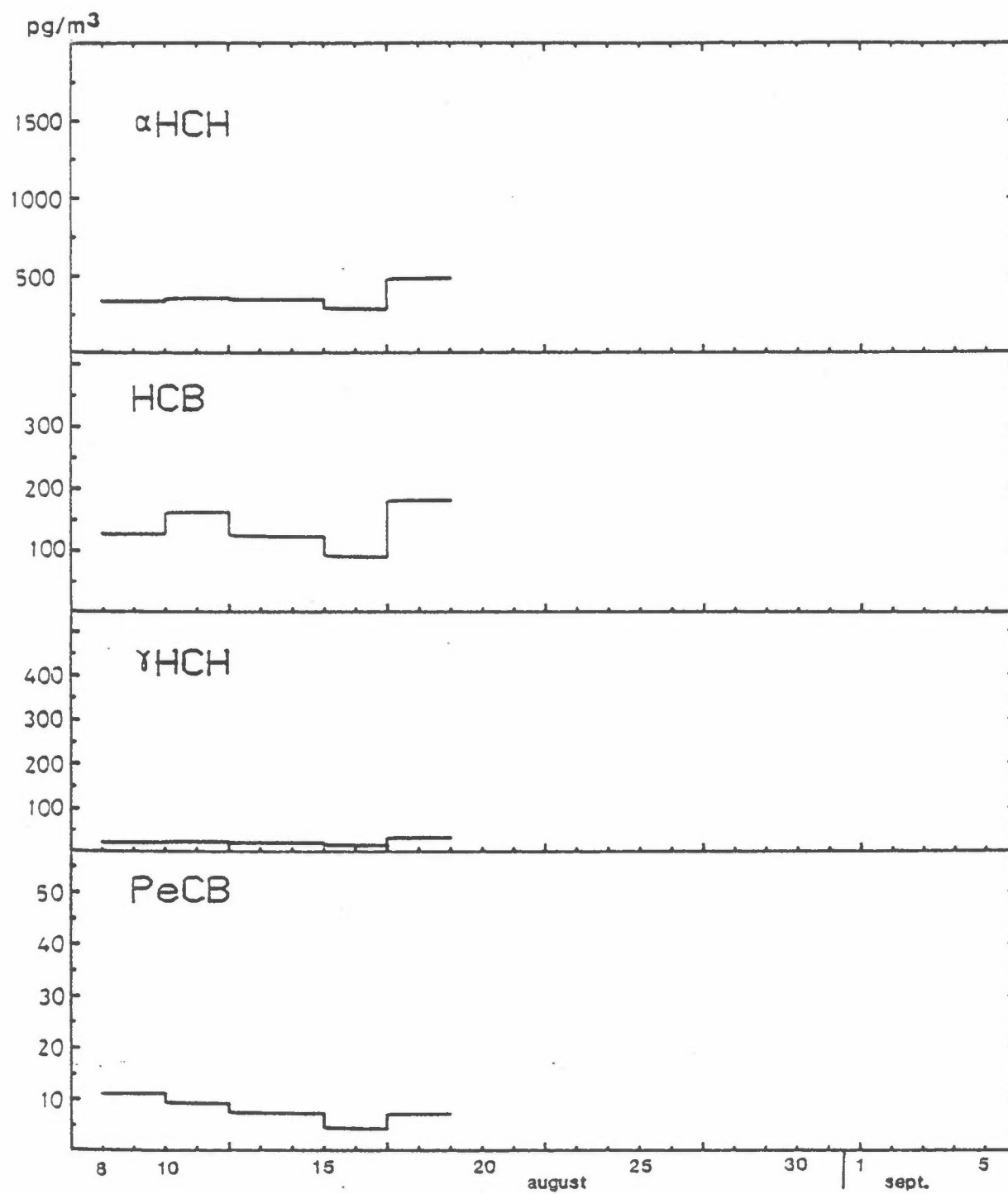


Figure 14: Time series of 2- and 3- day mean concentrations of hydrocarbons at Bjørnøya (BJO) during the Fall 1983 campaign.

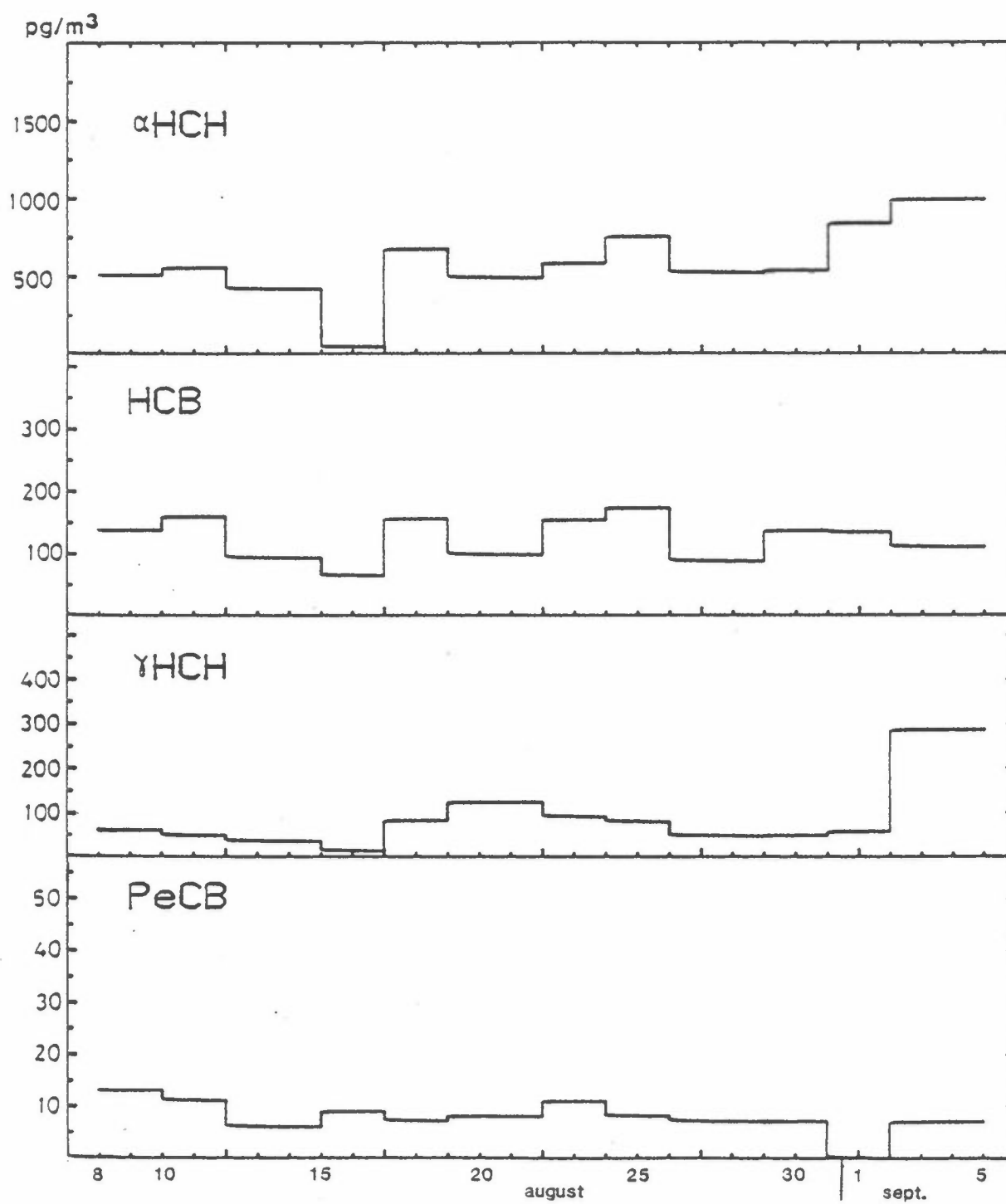


Figure 15: Time series of 2- and 3- day mean concentrations of hydrocarbons at Skrova (SKO) during the Fall 1983 campaign.

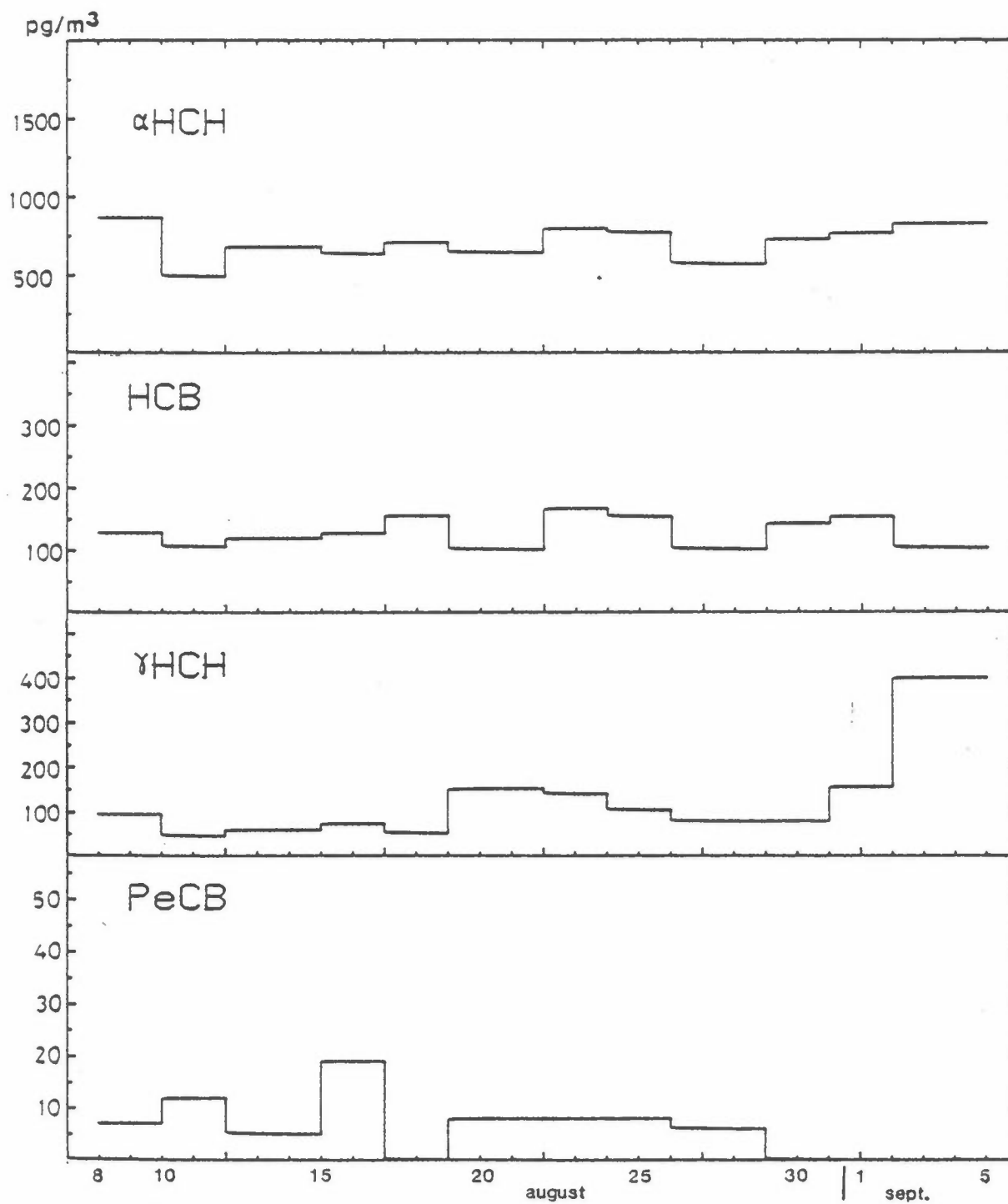


Figure 16: Time series of 2- and 3- day mean concentrations of hydrocarbons at Kårvatn (KAR) during the Fall 1983 campaign.

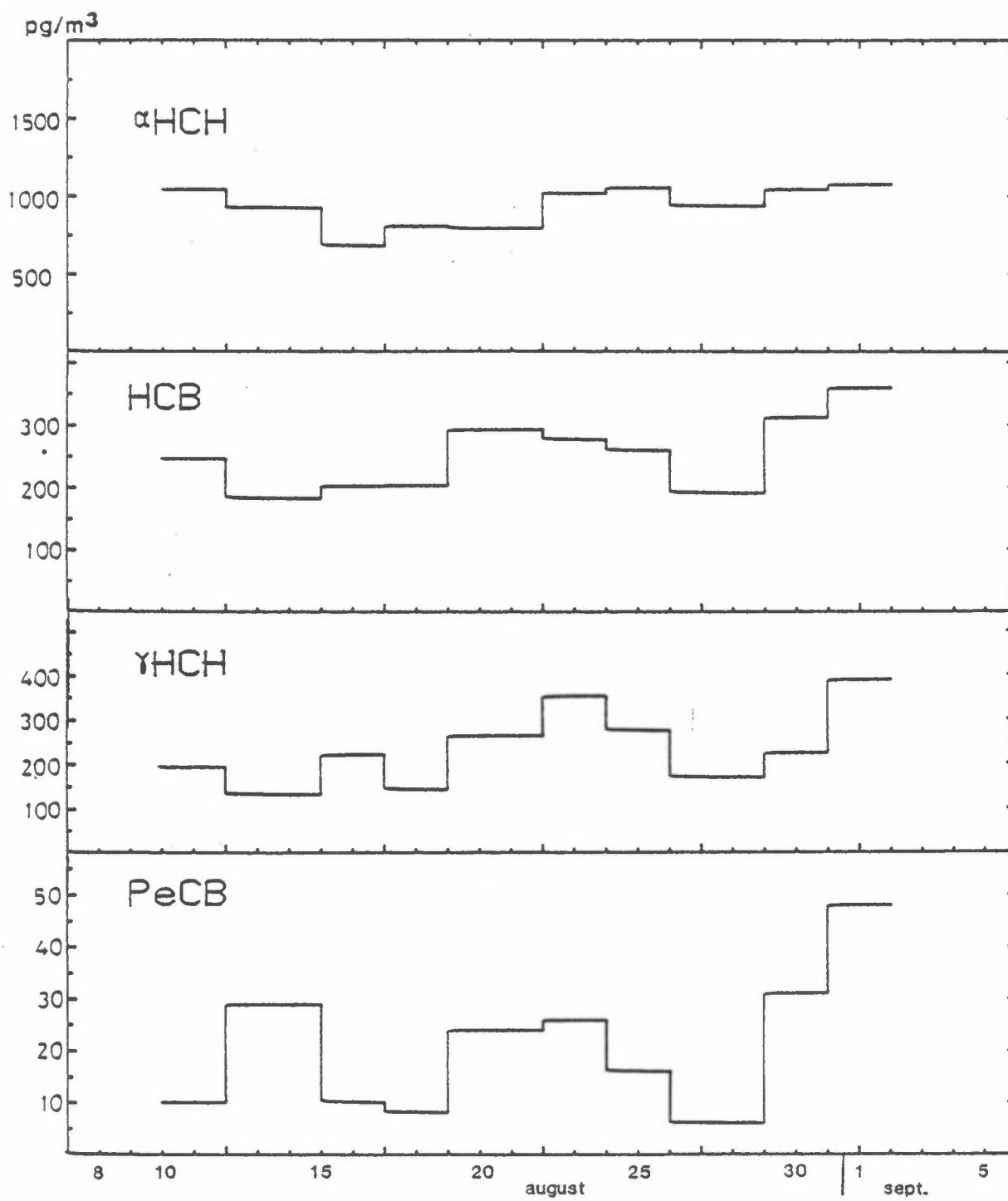


Figure 17: Time series of 2- and 3- day mean concentrations of hydrocarbons at Birkenes (BIR) during the Fall 1983 campaign.

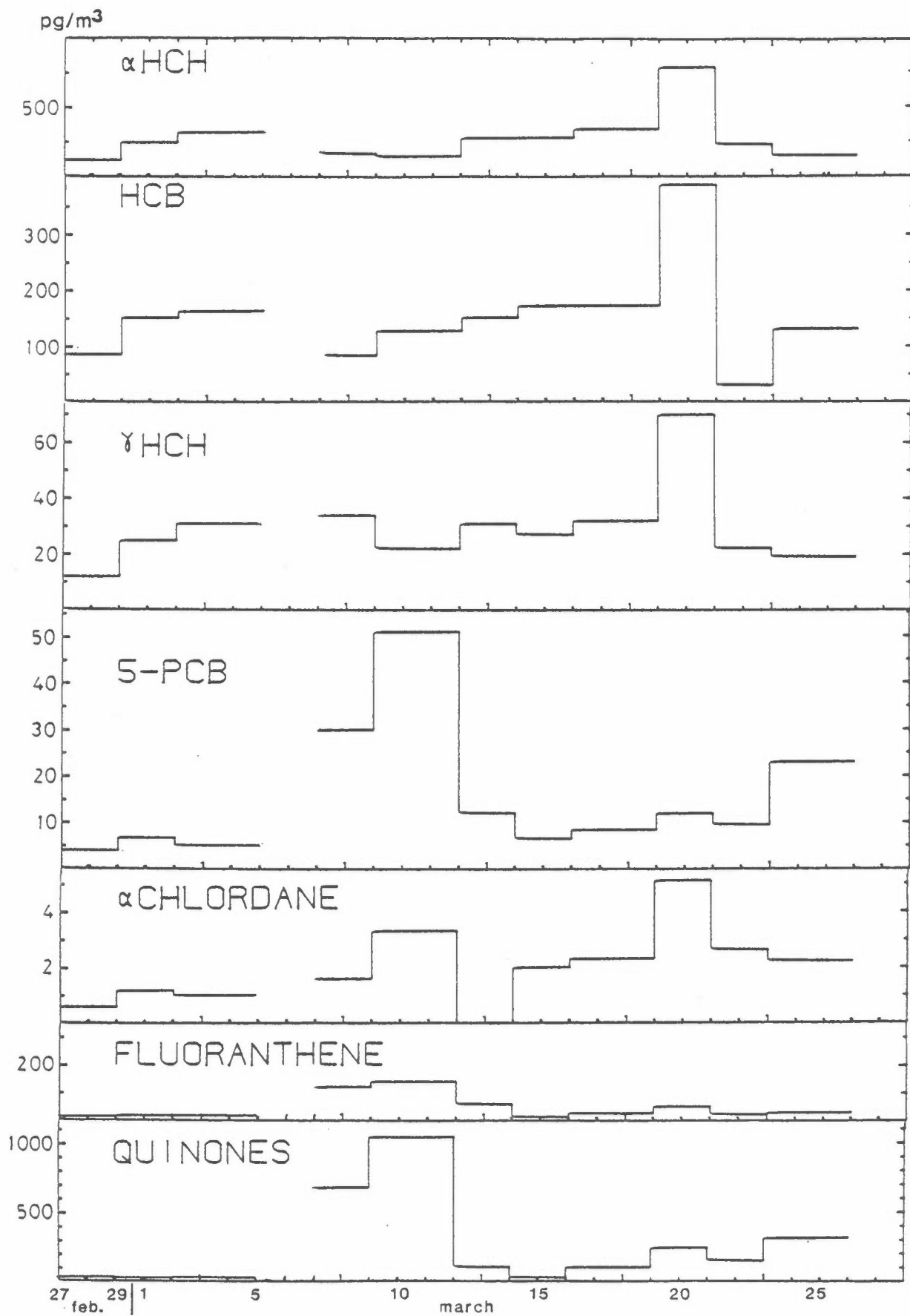


Figure 18: Time series of 2- and 3- day mean concentrations of hydrocarbons at Ny Alesund (NYA) during the Winter/Spring 1984 campaign.

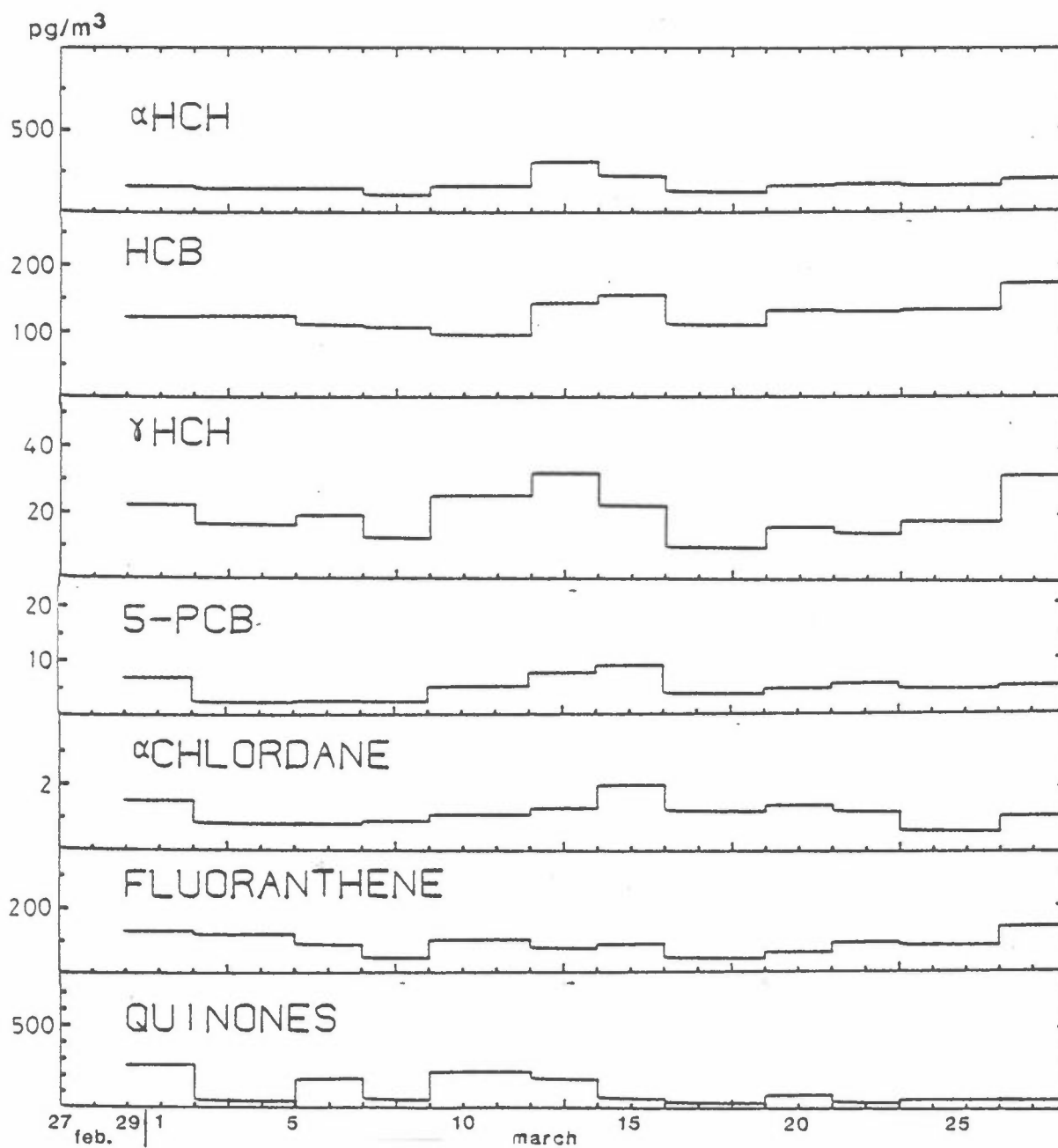


Figure 19: Time series of 2- and 3- day mean concentrations of hydrocarbons at Jergul (JER) during the Winter/Spring 1984 campaign.

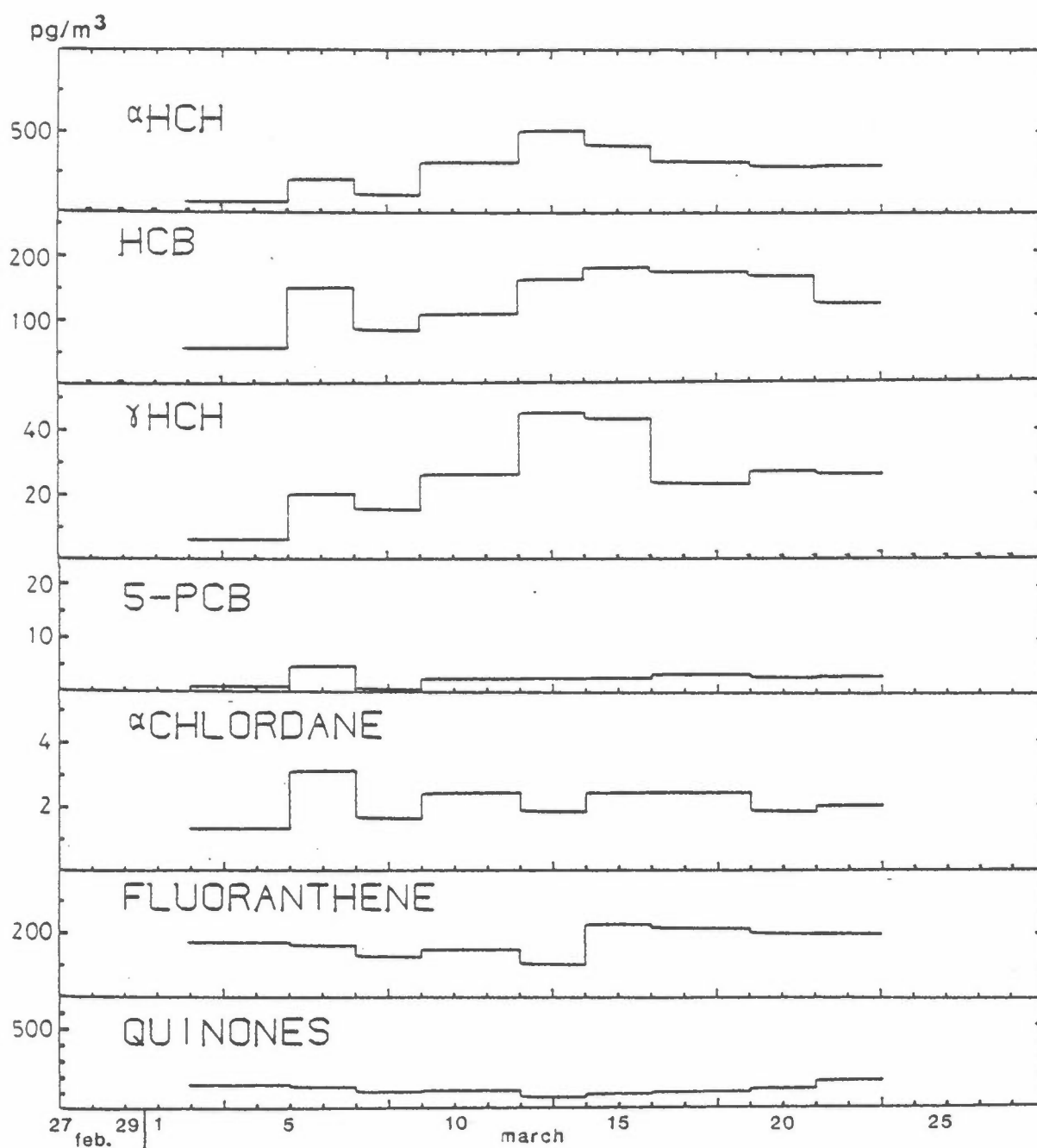


Figure 20: Time series of 2- and 3- day mean concentrations of hydrocarbons at Kárvatn (KAR) during the Winter/Spring 1984 campaign.

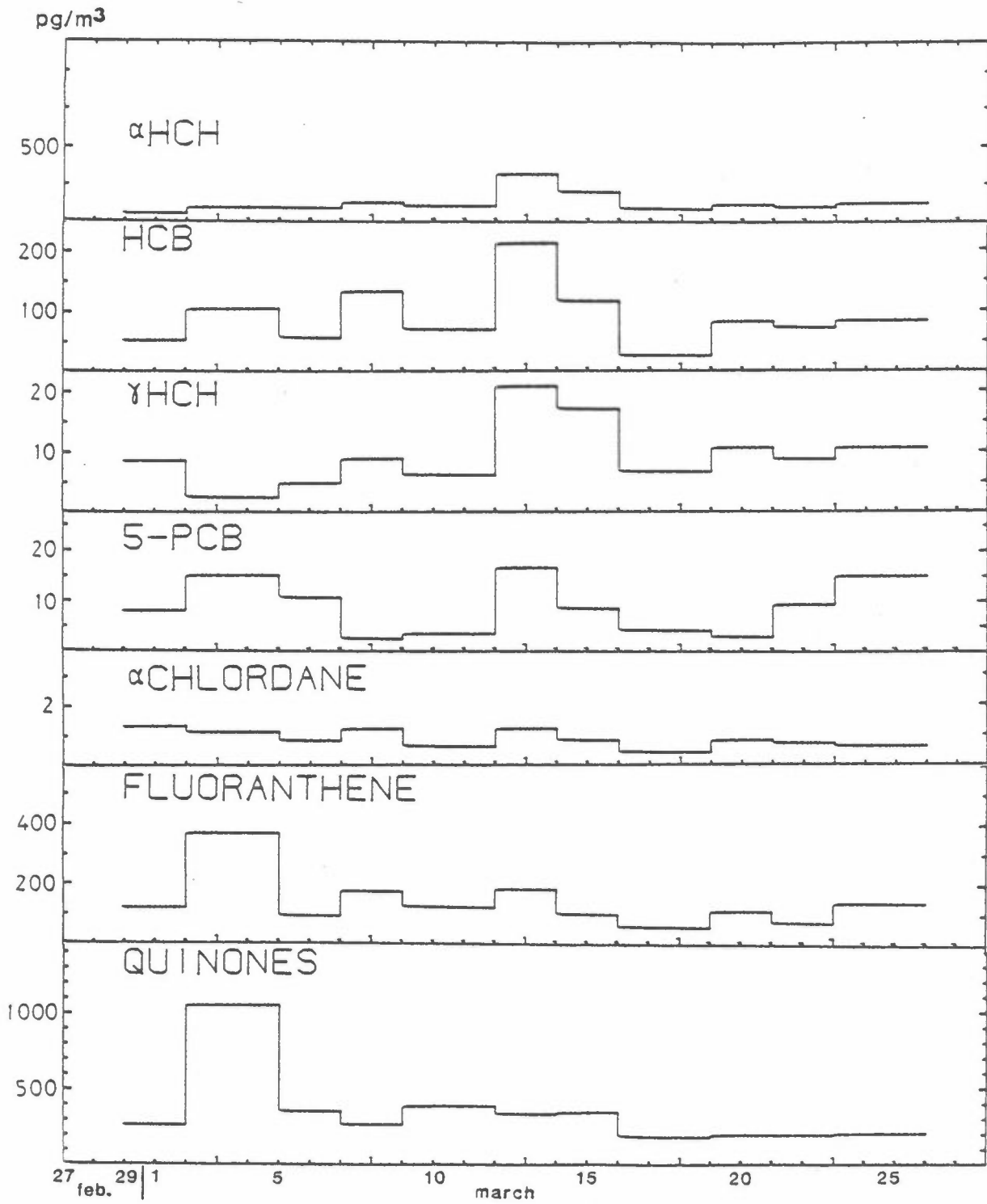


Figure 21: Time series of 2- and 3- day mean concentrations of hydrocarbons at Birkenes (BIR) during the Winter/Spring 1984 campaign.

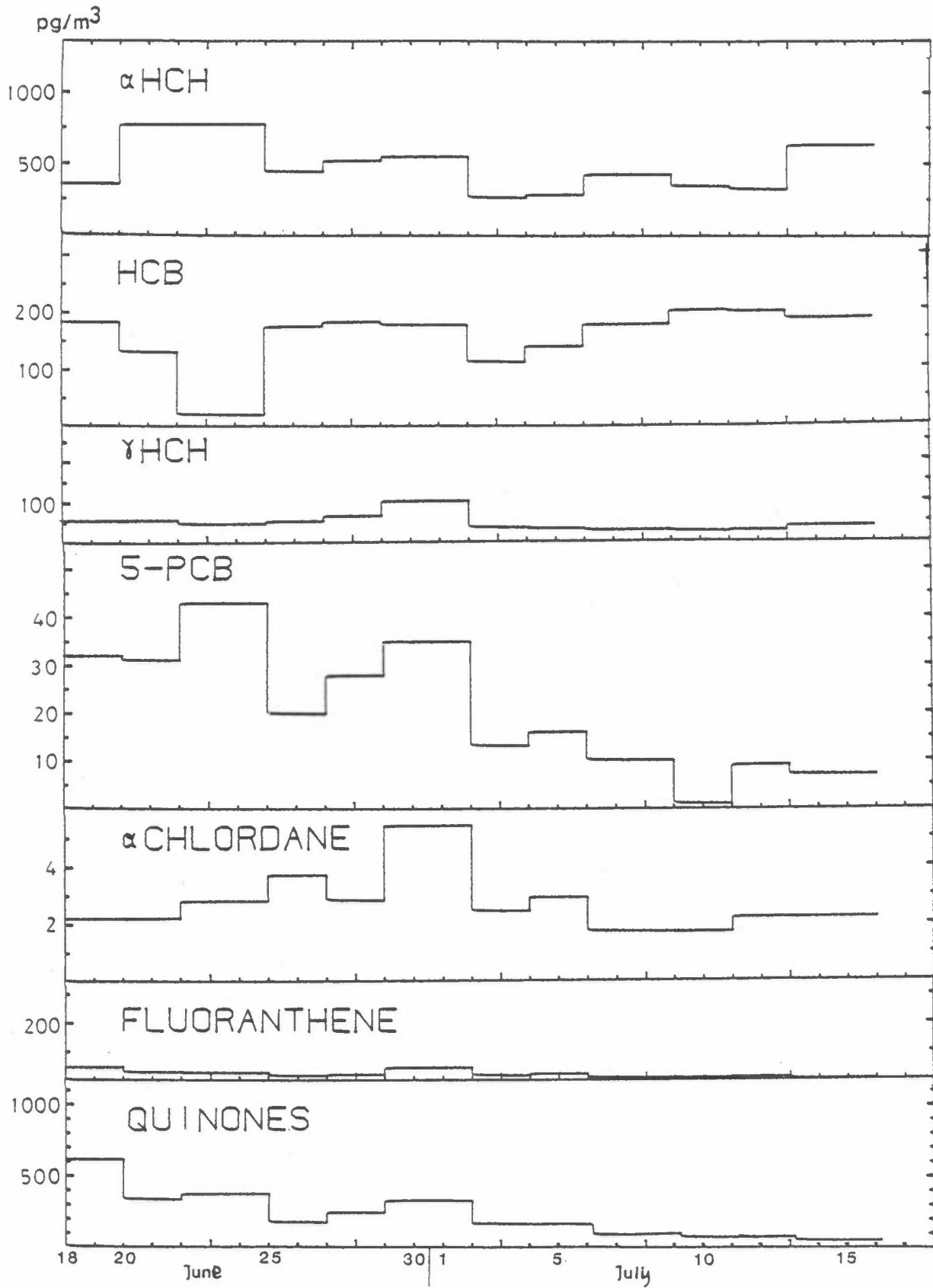


Figure 22: Time series of 2- and 3- day mean concentrations of hydrocarbons at Ny Alesund (NYA) during the Summer 1984 campaign.

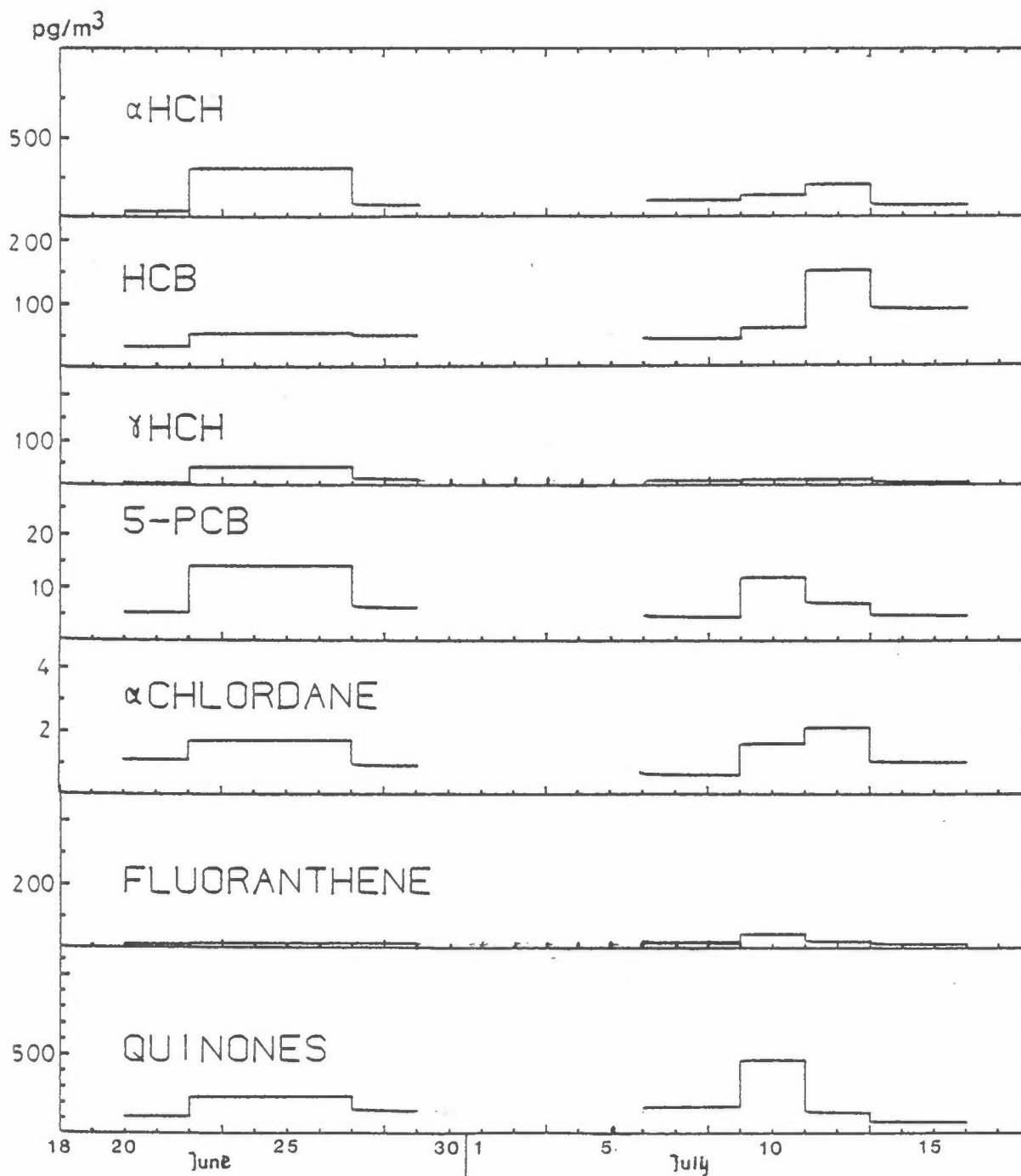


Figure 23: Time series of 2- and 3- day mean concentrations of hydrocarbons at Bjørnøya (BJO) during the Summer 1984 campaign.

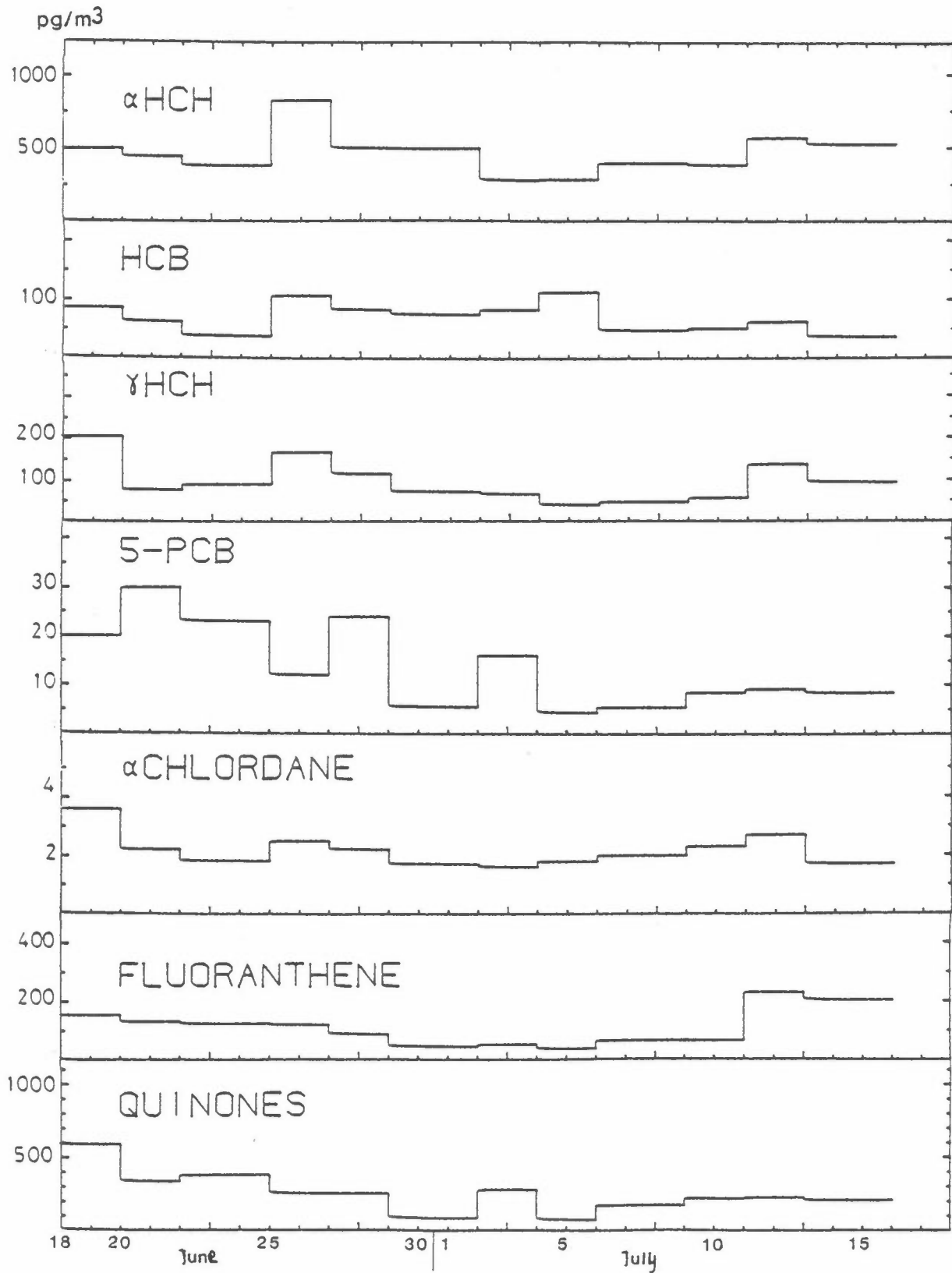


Figure 24: Time series of 2- and 3- day mean concentrations of hydrocarbons at Jergul (JER) during the Summer 1984 campaign.

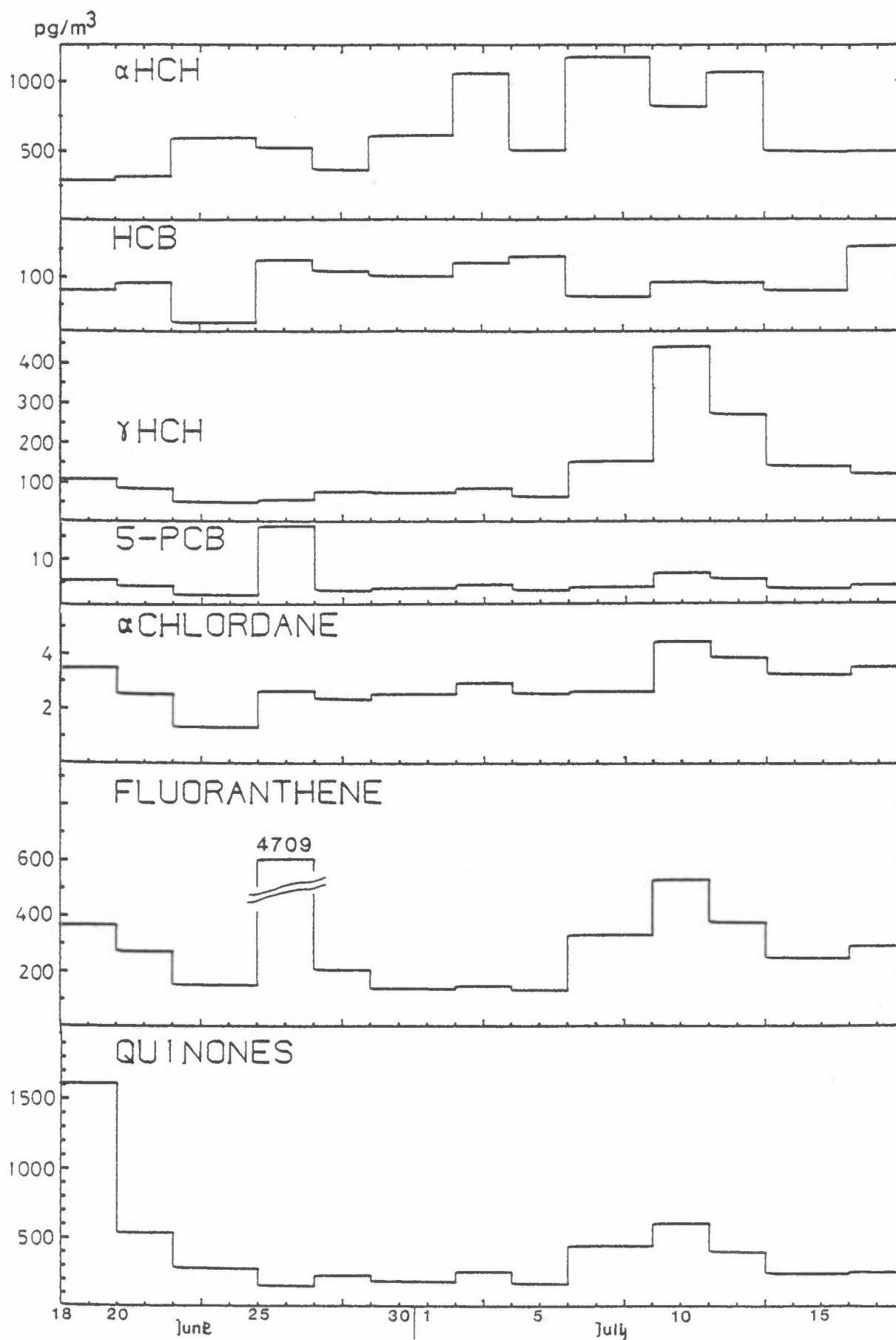


Figure 25: Time series of 2- and 3- day mean concentrations of hydrocarbons at Kårvatn (KAR) during the Summer 1984 campaign.

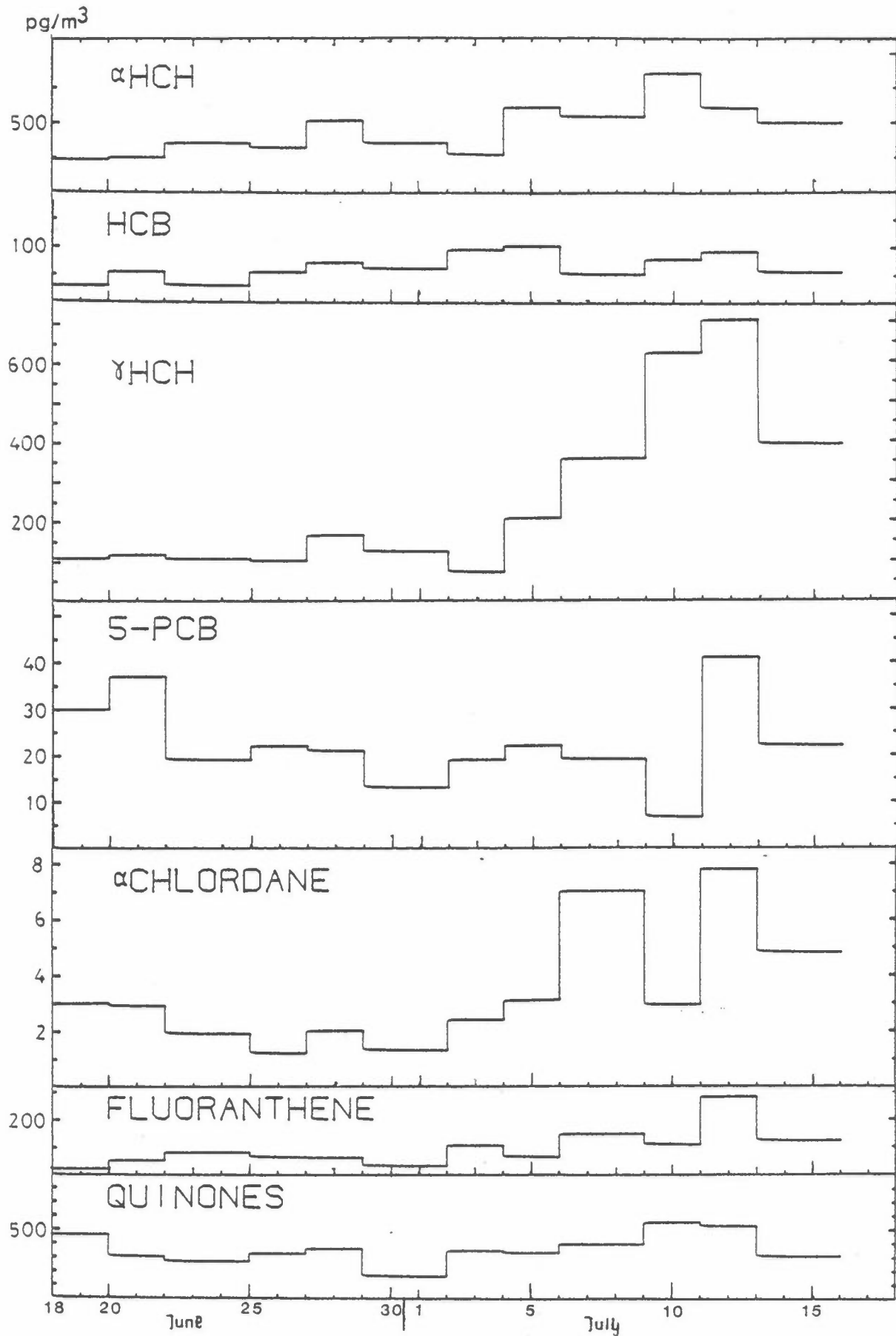


Figure 26: Time series of 2- and 3- day mean concentrations of hydrocarbons at Birkenes (BIR) during the Summer 1984 campaign.

Table 1: Periods for intensive measurement campaigns at all BP ground stations.

Station	1982	1983		1984	
	Fall	Winter/Spring	Fall	Winter/Spring	Summer
NYA	08-16 to 09-13	03-01 to 04-06	08-08 to 09-04	02-27 to 03-26	06-18 to 07-16
BJO	08-18 to 09-13	02-28 to 03-30	08-08 to 08-18		06-20 to 07-16
HOP	08-16 to 09-13	02-28 to 03-28			
JAM	08-16 to 09-13	02-28 to 03-28			
JER				02-29 to 03-28	06-18 to 07-16
SKO			08-08 to 09-05		
KAR			08-08 to 09-05	02-02 to 03-23	06-18 to 07-18
BIR	08-27 to 09-13		08-10 to 09-02	03-29 to 03-26	06-18 to 07-16

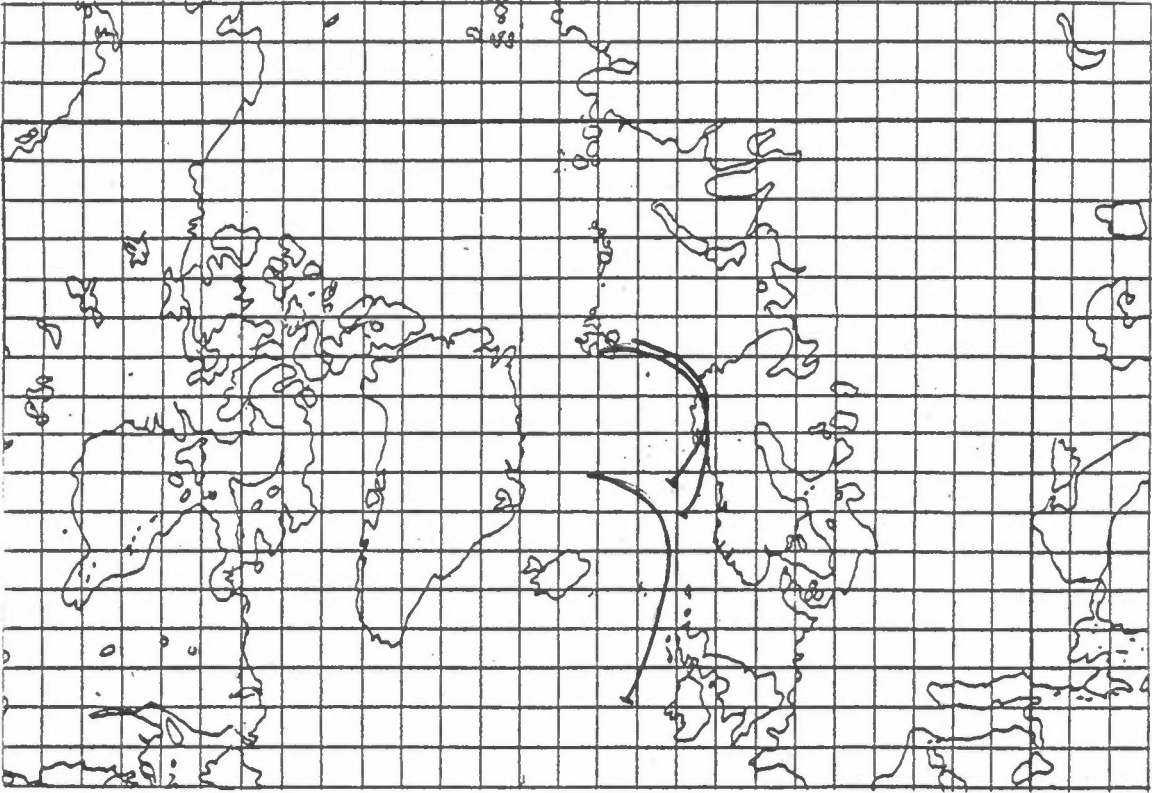
5 ORIGIN OF HYDROCARBONS IN THE ARCTIC AIR

5.1 AUGUST/SEPTEMBER 1982 CAMPAIGN

The two- and three day mean concentrations of hydrocarbons at NYA, HOP, BJO, JAM and BIR are summarized in Figures 4 through 8, respectively.

At the beginning of the campaign enhanced concentrations of γ -HCH, 5-PCB, α -chlordane, fluoranthene and quinones were observed at NYA and JAM. The 850 mb wind trajectories, shown in Figure 27, indicate that different air masses were measured at NYA and JAM. The former station was likely affected by air passing over the Kola Peninsula and Finland, while the latter station by Atlantic air passing over Central Scandinavia. It is interesting to observe the difference of the ratio of γ - to α -HCH isomers at NYA and JAM. Generally this ratio is very low, much lower than observed in Lillestrøm, Southern Norway (Oehme and Ottar, 1984). Low ratios of γ - to α -HCH in the North Atlantic air were also reported by Bidleman and Leonard (1982). They suggested the use of "BHC" pesticide mixtures as a possible explanation. Technical "BHC" contains about 55-80% α -HCH and 8-15% γ -HCH (Metcalf, 1955). However, they did not explain whether the high proportion of α - to γ -HCH results from the continued use of technical "BHC" mixtures (or other mixtures, e.g. technical HCH) or to isomerization of γ -HCH to α -HCH in the environment. The γ - to α -ratios observed at NYA and JAM from 16 to 17 August 1982, the highest during the Fall 1982 measurement campaign, were below 1:100. Since the content of γ -HCH in pesticides used in Norway is more than

1982-08-16



1982-08-17

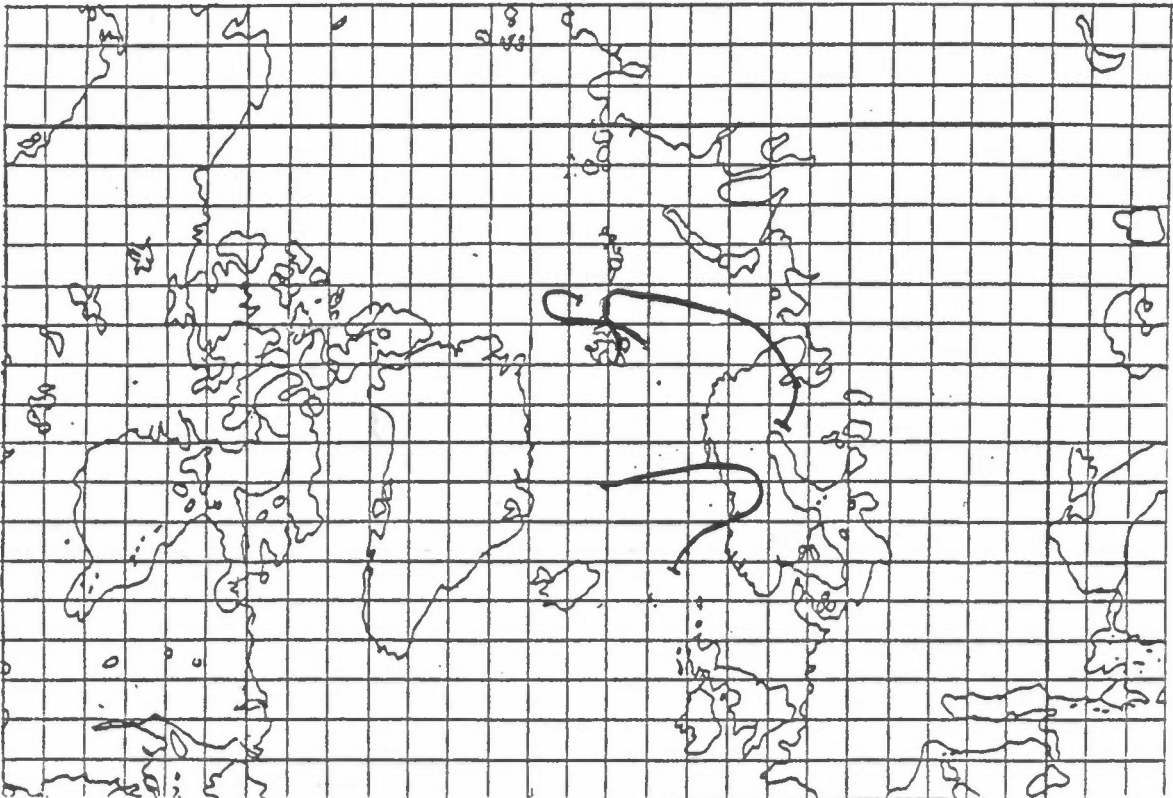


Figure 27: Daily 850 mb trajectories for NYA, HOP and JAM during the period 1982-08-16 to 1982-08-17.

99%, sources in Central Scandinavia do not seem to contribute to enhanced concentrations of polychlorinated hydrocarbons at JAM during August 1982. Although unknown, the sources may be the same as those resulting in the low γ - to α -HCH ratio in the Atlantic air observed by Bidleman and Leonard (1982).

In the case of the HCH concentrations at NYA, the agricultural application of HCH in the USSR and Finland may be considered as a potential source. The γ - to α -HCH ratio in pesticides used in the USSR is unknown. The forest pesticides in Finland contain ca. 20% of the γ -HCH-isomer (Tiittanen and Blomquist, 1982).

The enhanced concentrations of fluoranthene at both stations can be related to emissions from gasoline combustion in Scandinavia and the Kola Peninsula. Increased concentrations of Pb at the same time at NYA and JAM (Pacyna et al., 1985a) seem to confirm this hypothesis. Local emissions from fuel combustion should also be considered as a potential contributor to the enhanced concentrations of fluoranthene at that time.

The very high concentrations of quinones at NYA and JAM are probably due to emissions from fuel combustion in Scandinavia and the Kola Peninsula, as well as on Spitsbergen. The other explanation is that quinones may be decomposition products of biological materials. Some doubts about this hypothesis arise when considering that very high concentrations of quinones are observed at NYA and JAM at the same time and appear no more during the Fall 1982 campaign.

The slightly enhanced concentrations of γ -HCH at NYA and quinones at HOP during the periods August 23-26, and August 29 to September 1, 1982 can be related to air mass transport over the northern Soviet Union and the Arctic Ocean (see 850 mb trajectories in Figures 28 and 29). A similar explanation was suggested to explain enhanced concentrations of trace elements and some ions at both stations during the same period (Pacyna et al., 1985a).

The next increase of pollutant concentrations was observed between September 1 and September 3, 1982 at HOP and BJO. The 850 mb trajectories (Figure 30) indicate transport of air masses across Scandinavia, while clean air masses, circulating over the Arctic Ocean prevailed over NYA and JAM.

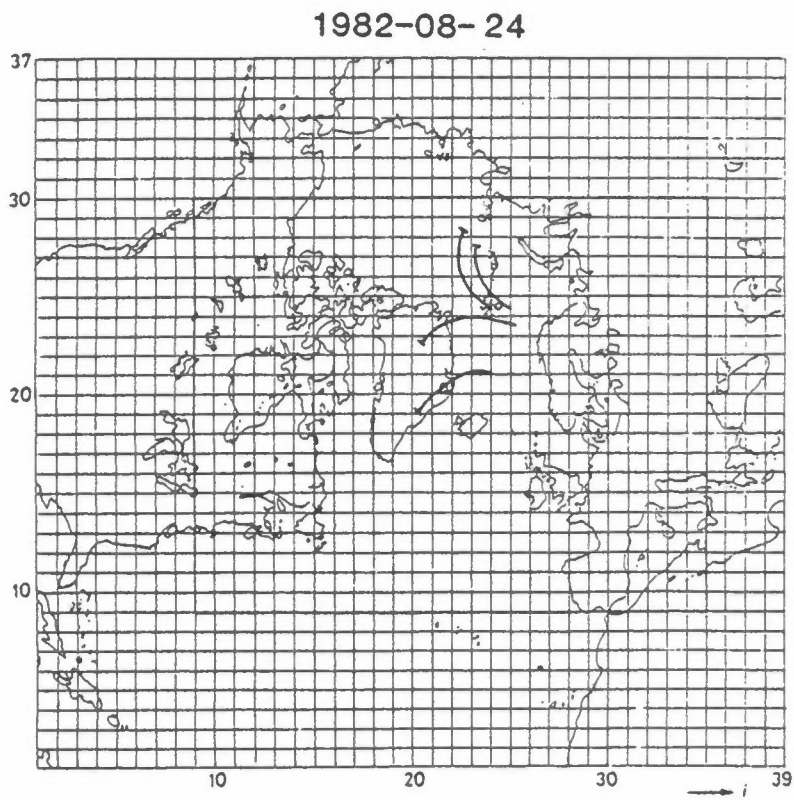
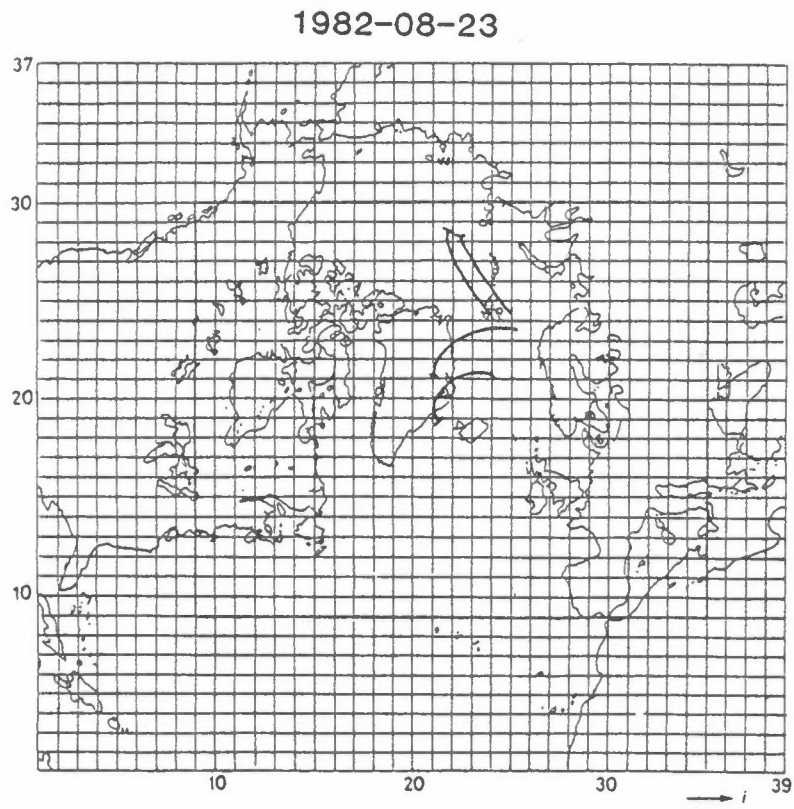
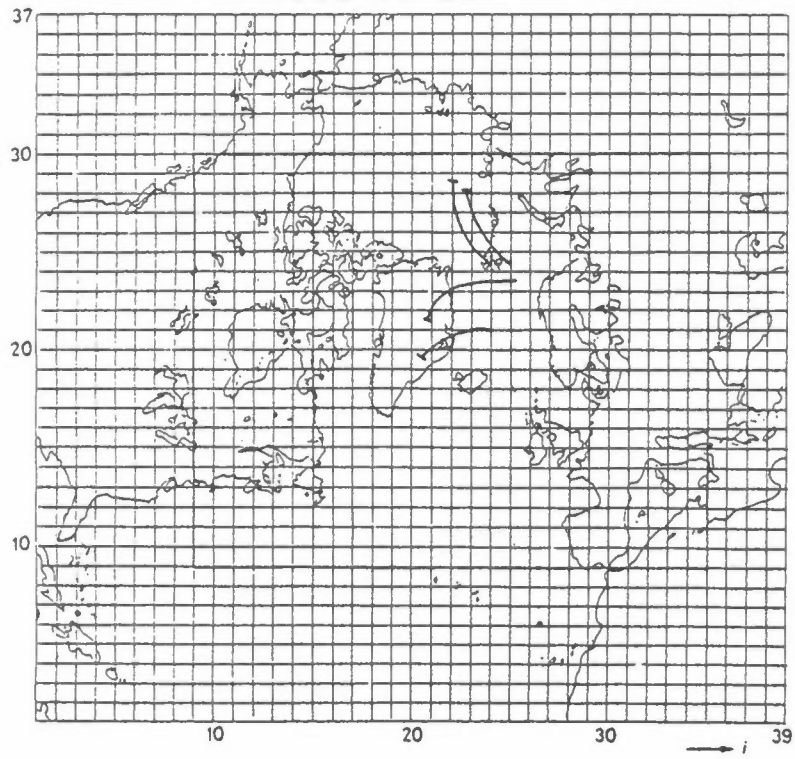


Figure 28: Daily 850 mb trajectories for JAM, BJO, HOP and NYA during the period 1982-08-23 to 1982-08-26.

1982-08-25



1982-08-26

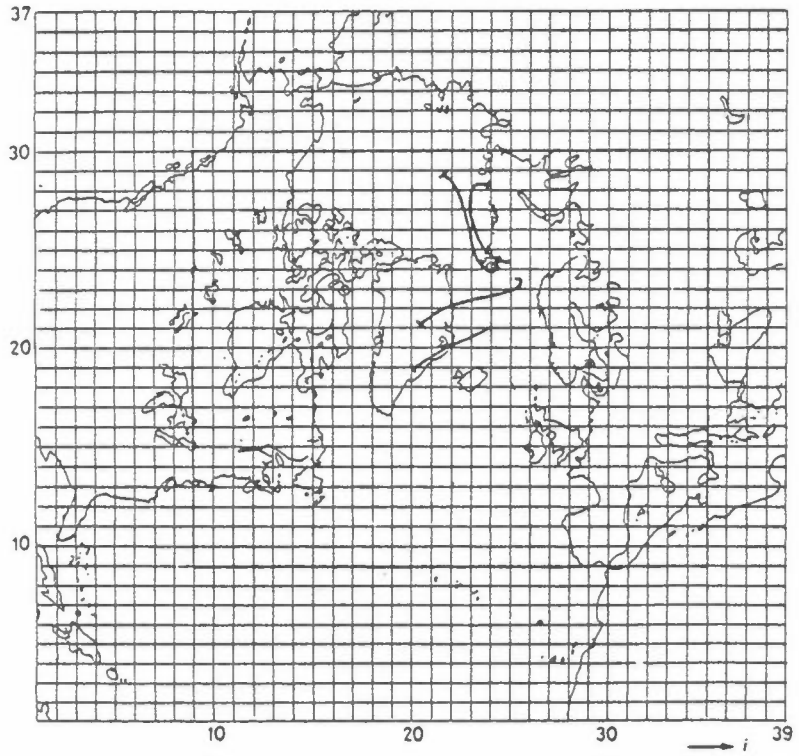


Figure 28: Cont.

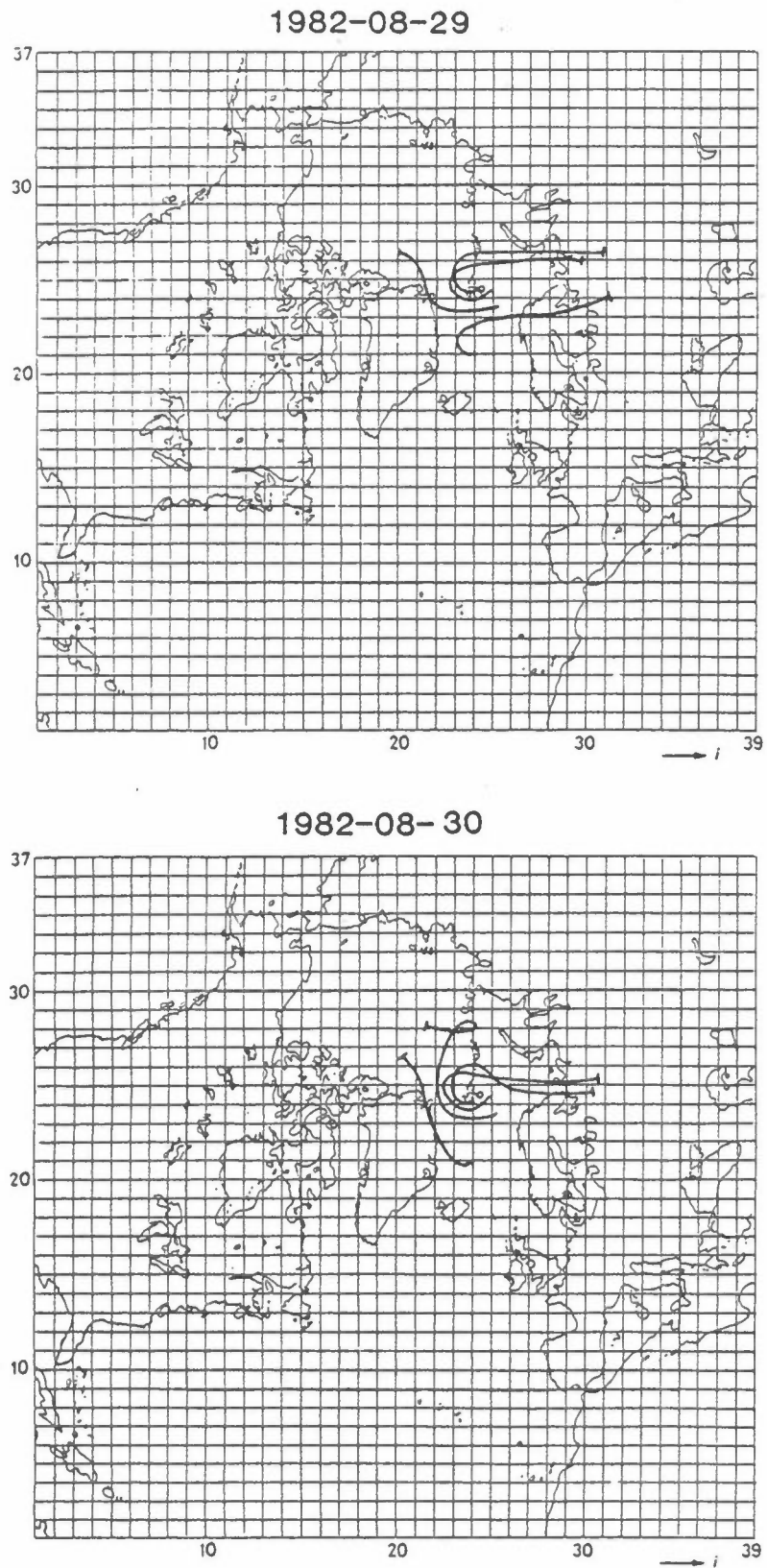


Figure 29: Daily 850 mb trajectories for JAM, BJO, HOP and NYA during the period 1982-08-29 to 1982-09-01.

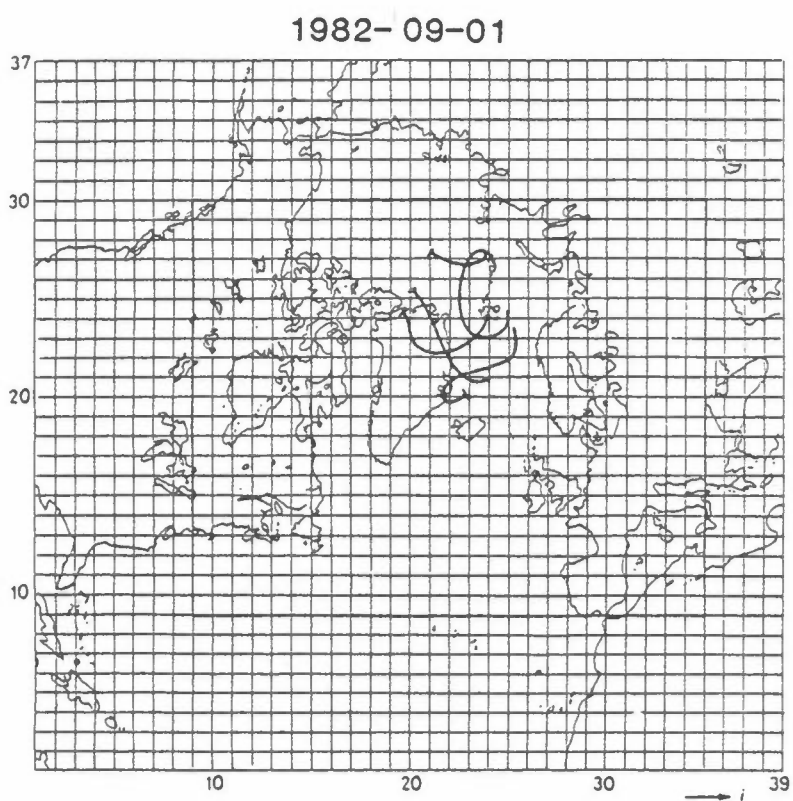
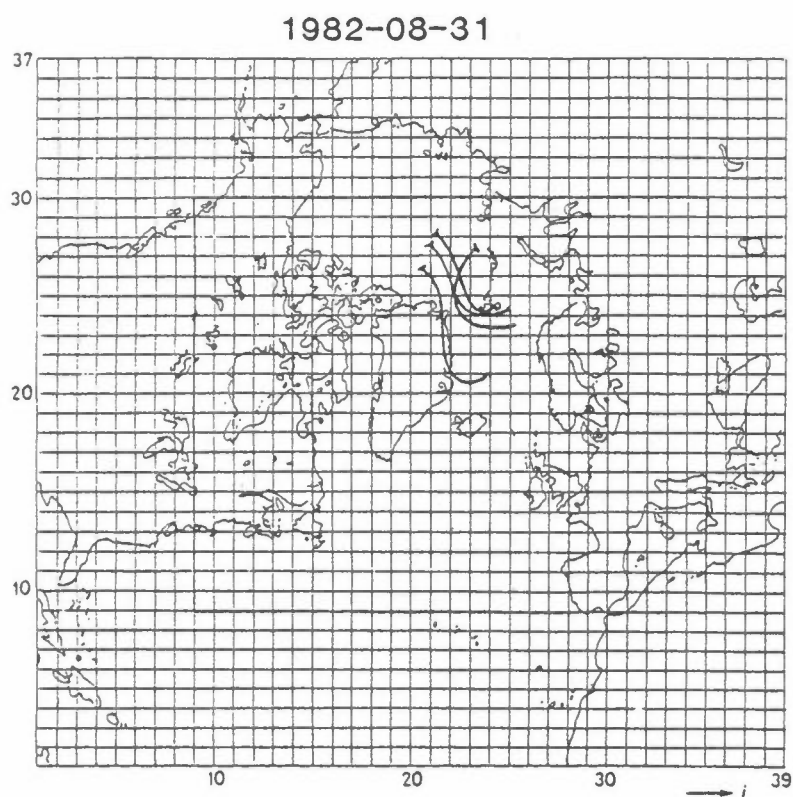


Figure 29: Cont.

1982-09-02

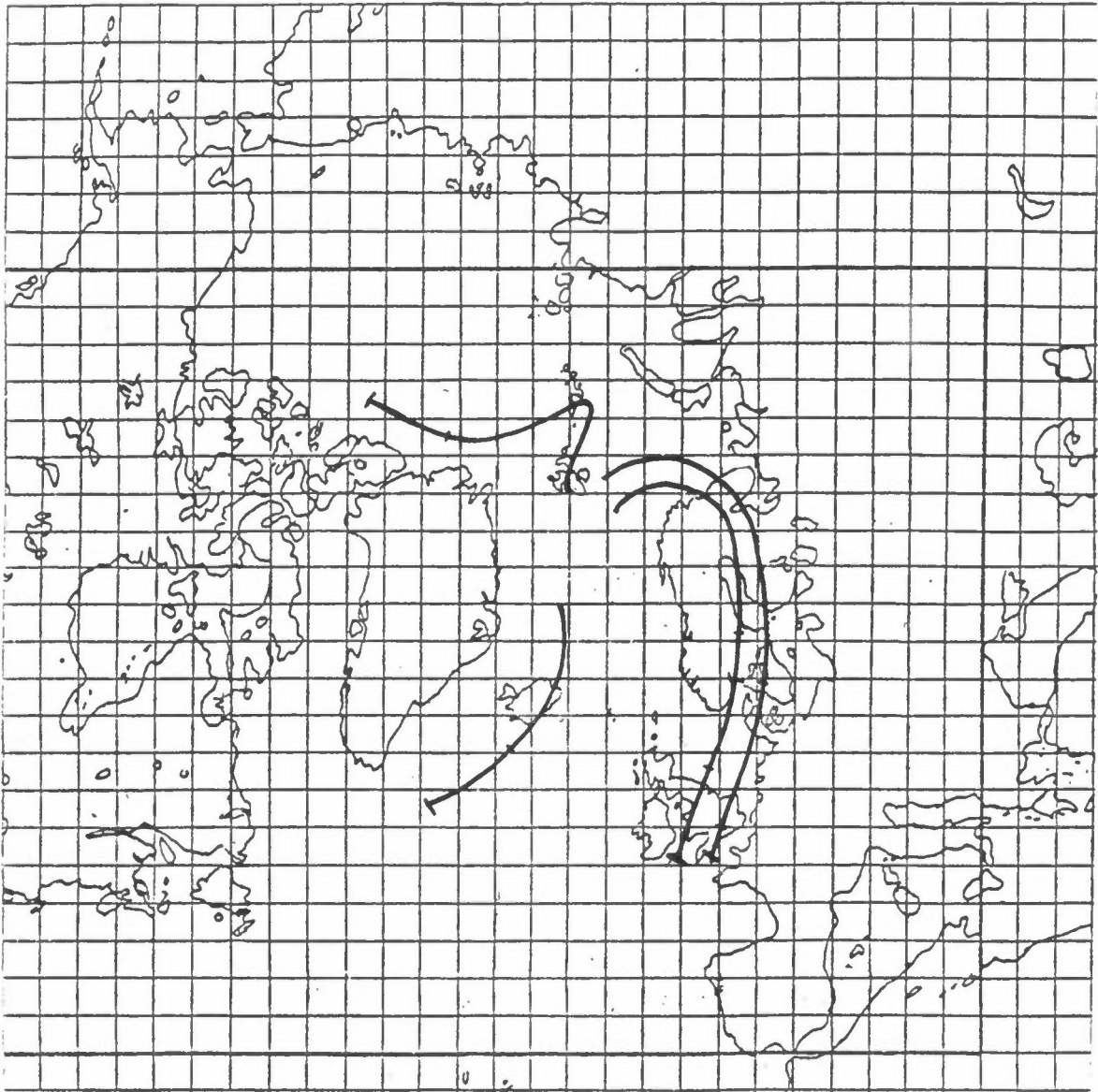


Figure 30: Daily 850 mb trajectories for JAM, BJO, HOP and NYA during the period 1982-09-02.

The concentrations of α - and γ -HCH at HOP and BJO were high again between September 6 and September 9, 1982. The 850 mb wind trajectories, shown in Figure 31, now indicate a transport of arctic air across northern Greenland to all four arctic stations. On this basis the air masses could have originated on the North American continent. If so, all four stations would be expected to show similar concentrations of HCH which is not the case. Also, no inorganic components can be found to support a North American origin of the pollution measured in the Norwegian Arctic at that time (see Pacyna et al., 1985a).

A second explanation could be re-emission of previously deposited material at HOP and BJO. However, other hydrocarbons do not indicate any trace of such re-emission at these two stations during the period September 6-September 9, 1982. A third possibility is vertical exchange of aerosols under certain meteorological conditions. It has been shown that transport of air masses over several thousand kilometers may affect the quality of the arctic air both summer and winter (Pacyna et al., 1985b). Polluted air layers, carrying a mixture of anthropogenic and natural air pollutants from a variety of sources in different geographical areas have been identified in the arctic atmosphere at altitudes from 2.0 km to 4-5 km. No evidence exists in the literature on the content of chlorinated hydrocarbons in these layers. However, they may contain HCH isomers, as their origin can be traced back to Asia (Pacyna and Ottar, 1986), the area with the larger use of pesticides. The atmospheric residence time of HCH is a few months, and in summer, small particles containing HCH among other pollutants may coagulate more efficiently than in winter, and be deposited.

The data from BIR do not indicate any increase of the hydrocarbon concentrations discussed in this report.

5.2 MARCH/APRIL 1983 CAMPAIGN

The two- and three-day mean concentrations are summarized in the time-series given in Figures 9-12 for NYA, HOP, BJO and JAM, respectively. There were well-defined concentration peaks of α -HCH, and γ -HCH at NYA and HOP between March 11 and March 13, 1983. Concentration peaks of γ -HCH, 5-PCB, fluoranthene and quinones were also observed at JAM.

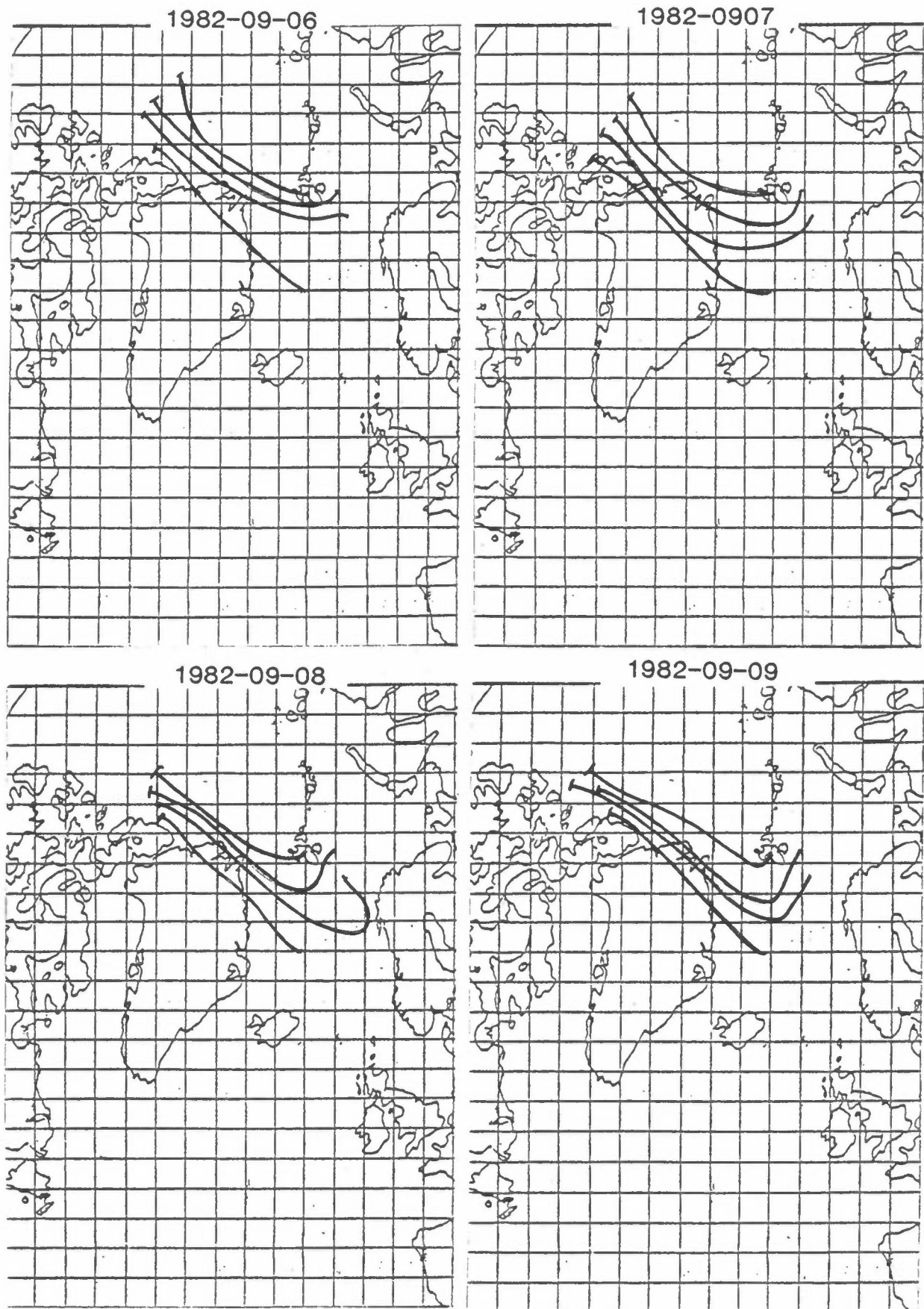


Figure 31: Daily 850 mb trajectories for JAM, BJO, HOP and NYA during the period 1982-09-06 to 1982-09-09.

In Figure 32 the 850 mb wind trajectories to NYA (Harris, 1984) are reproduced. They indicate that the main sources of these pollutant hydrocarbons may be located on the Kola Peninsula and along the Urals in the USSR. The ratio of γ - to α -HCH at NYA was 1:14, 1:18 at HOP, and 1:13 at JAM. These values are significantly higher than the γ - to α -HCH ratio obtained for August/September 1982, indicating that fresh aerosols were measured at NYA, HOP and JAM in March 1983. The high values of the γ - to α -HCH in March 1983 are due to high concentrations of γ -HCH isomer. This is rather surprising, because the γ -HCH is used as an insecticide in the spring-summer season. Therefore the higher concentrations in the Arctic would be expected in summer rather than in winter. No evidence was found in the literature on the efficiency of the γ - to α -HCH isomerization. Our results seem to suggest that the isomerization of γ - to α -HCH during the air mass transport to the Arctic is significantly faster in summer than in winter.

The enhanced concentrations of hydrocarbons at NYA, HOP and BJO were observed again between March 21 and March 25, 1983. The daily 850 mb trajectories for NYA, shown in Figure 33, indicate again that the air masses passed over the USSR. The γ - to α -HCH ratio was similar to that obtained during the previous episode of long-range transport of air pollutants from the USSR to the Norwegian Arctic.

The concentration peaks at JAM appeared between March 23 and March 25, 1983. The apparent transport time for the pollutants between NYA, HOP and BJO, and JAM was 2 days. It can be speculated, that the polluted air masses reached NYA first, and then turned over Spitsbergen to JAM.

The suggested origins of the March 1983 episodes in the Norwegian Arctic resulting in concentration peaks of various hydrocarbons, are in very good agreement with the findings of the studies on behaviour of trace elements and ions in the Arctic (Pacyna et al., 1985a) during the same period.

5.3 AUGUST/SEPTEMBER 1983 CAMPAIGN

The time series of two- and three day mean concentrations of α - and γ -HCH, HCB and PeCB at NYA, BJO, SKO, KAR and BIR in August/September 1983 are shown in Figures 13 through 17, respectively. The data for BJO (Figure 14)

1983-03-11/13

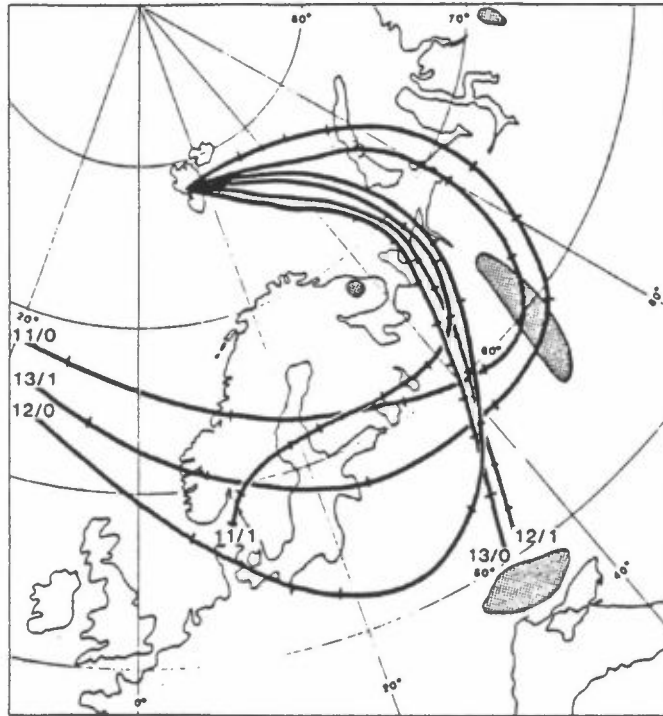


Figure 32: Daily 850 mb trajectories to NYA during the period 1983-03-11 to 1982-03-13, at 00 00 UT (indicated by 0 after the slash) and 1200 UT (1 after slash). Daily marks are provided on each trajectory. The shaded areas represent main emission regions in the USSR.

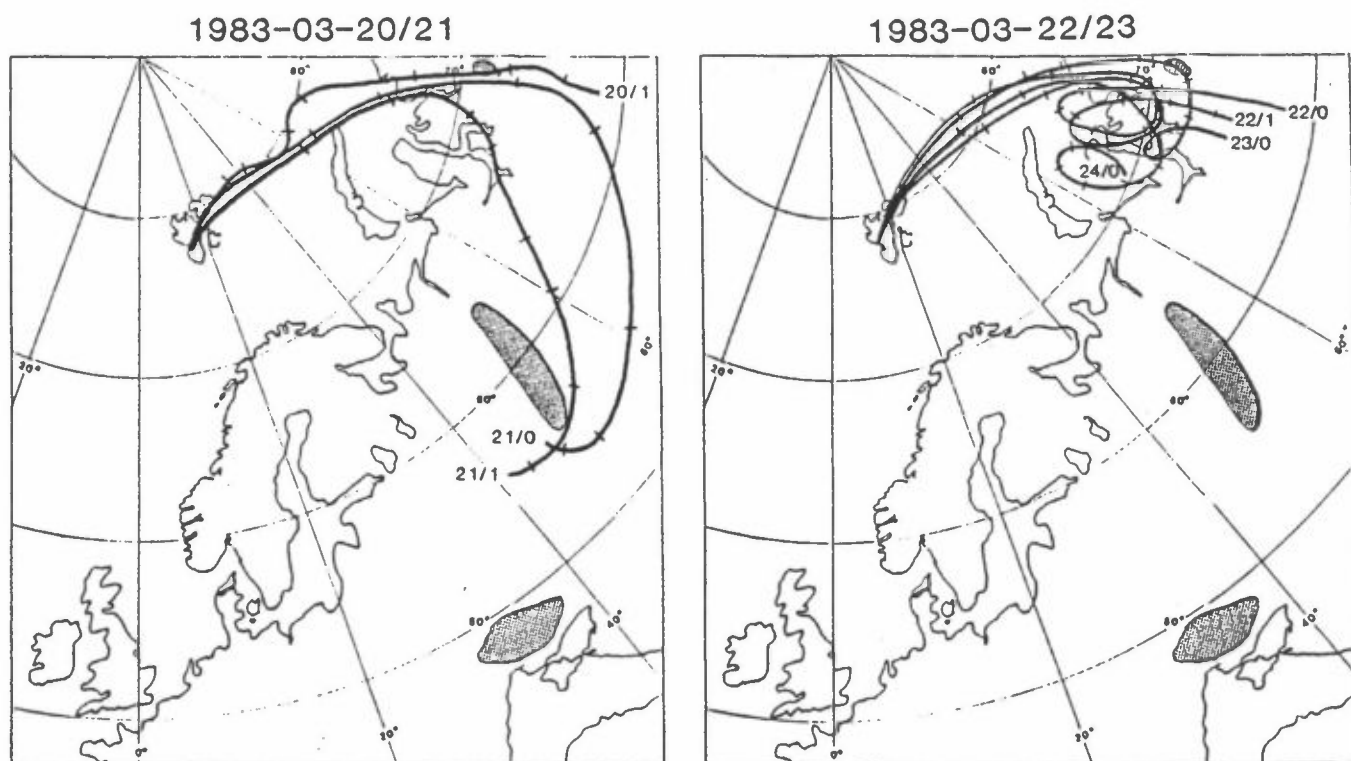


Figure 33: Daily 850 mb trajectories to NYA during the period 1983-03-20 to 1983-03-23, at 00 00 UT (indicated by 0 after the slash) and 1200 UT (1 after slash). Daily marks are provided on each trajectory. The shaded areas represent main emission regions in the USSR.

are incomplete due to a failure of the sampler. The α - and γ -HCH and HCB concentrations were rather constant at NYA. The data do not indicate episodes of long-range transport of polluted air to the Arctic at that time (Pacyna et al., 1985a; Vitols and Pacyna, 1985). The γ -HCH concentrations at NYA were higher in 1983 than in the same period of 1982 by at least one order of magnitude. At the same time the α -HCH concentrations were lower, resulting in a high γ - to α -HCH ratio for the August/September 1983 campaign. The ratio is about 1:10 and an intriguing question is what might have been the source of the high γ -HCH concentrations at NYA assuming no direct contribution of long-range transport. One possible explanation is re-emission of γ -HCH from the sea. Due to its several unusual features, the period between 14-24 August 1983 at NYA has been examined in detail by Vitols and Pacyna (1985). High peaks in the 12-h condensation nuclei concentration medians occurred on several occasions, while the particle scattering coefficients (σ_{sp}) remained sub-detectable at the same time, suggesting the presence of fresh nucleation mode (or Aitken range) particles, possibly the result of relatively local gas-to-particle conversion from biogenic emissions over the sea. If so, secondary emission of pesticides from the sea is also possible. Surface waters are a major sink for the pesticides which may accumulate in the surface film. As mentioned in section 2 of this report, Lewis and Lee (1976) reported, that the major input of pesticides to the ocean is via the atmosphere. This is particularly true in the spring-summer seasons, when most of the pesticides are used. There is no evidence in the literature, however, that γ -HCH can be transported with the sea currents to the Arctic and evaporated there. Thus, this hypothesis should be considered with caution.

Two well defined peaks of PeCB concentrations at NYA in August 1983 are likely due to sublimation of previously deposited material.

Measurements at the stations on mainland Norway indicate very high concentrations of γ -HCH at the beginning of September 1983. Weather maps for the 700 and 500/ 1000 mb levels (Europäischer Wetterbericht, 1983) indicated long-range transport of air pollutants from England and western Europe across Scandinavia to the Norwegian Arctic. The γ -HCH to α -HCH ratios of 1:2.5 at BIR, 1:2 at KAR and 1:3 at SKO are the result of high γ -HCH concentrations in the transported air. These high γ -HCH concentrations are due to the application of lindane in western Europe.

The above described long-range transport of polluted air from western Europe was traced up to the Norwegian Arctic, as shown by the high concentrations of several inorganic air pollutants at NYA and HOP (e.g. Pacyna et al., 1985a). Unfortunately, the organic pollutants were not sampled there in September 1983.

5.4 FEBRUARY/MARCH/APRIL 1984 CAMPAIGN

The two- and three-day mean concentrations for this campaign are summarized in Figures 18-21 for NYA, JER, KAR and BIR. Two periods with high concentrations of hydrocarbons were observed at NYA, from March 9 to March 11 and March 19 to March 20 1984. High concentrations of 5-PCB, α -chlordane, fluoranthene and quinones were observed during the first period and the concentrations of α - and γ -HCH, HCB and α -chlordane peaked during the second period. The 850 mb wind trajectories to NYA during on March 9-11, 1984 in Figure 34 indicate transport of clean Atlantic air to the Arctic. No increase of concentrations of inorganic air pollutants was observed (Pacyna et al. 1985a) at that time in the Norwegian Arctic. Thus, the high concentrations of fluoranthene and quinones were likely due to local combustion processes on Spitsbergen.

The 850 mb trajectories to NYA for the period March 19-20, 1984 are shown in Figure 35. They indicate transport of air masses from North America to the Norwegian Arctic. The γ - to α -HCH ratio was 1:10, smaller than for Western Europe, likely due to isomerization of γ -HCH to α -HCH.

The "Russian" episodes in the Winter 1984 measurement campaign (Pacyna et al. 1985a), examined on the basis of the chemical composition of the NYA aerosol and wind trajectories, are not easy to trace in Figure 18. However, the γ - to α -HCH ratio of 1:10 for the periods February 29 to March 1, 1984 and March 14-15, 1984 and the ratio of 1:13 observed March 23-25, 1984 are very close to the γ - to α -HCH ratio for the "Russian" episodes in the March/April 1983 campaign (see section 5.2).

There were several instances of elevated concentrations of hydrocarbons at the Norwegian mainland stations. Enhanced concentrations were observed during the periods March 2-4 and March 7-8 at BIR, and during the period

1984-03-09 to 1984-03-11

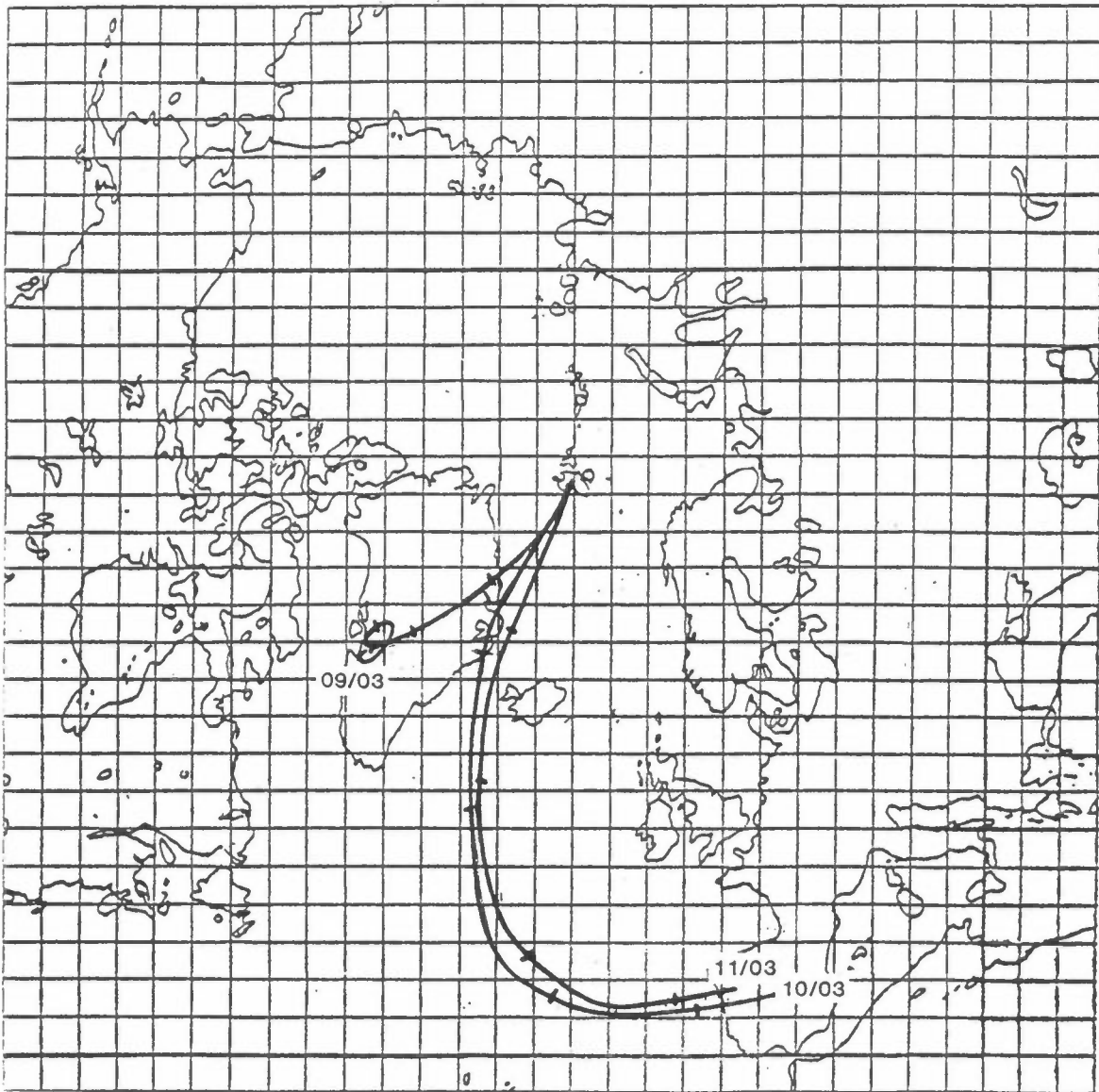


Figure 34: Daily 850 mb trajectories to NYA during the period 1984-03-09 to 1984-03-11.

1984-03-19 to 1984-03-20

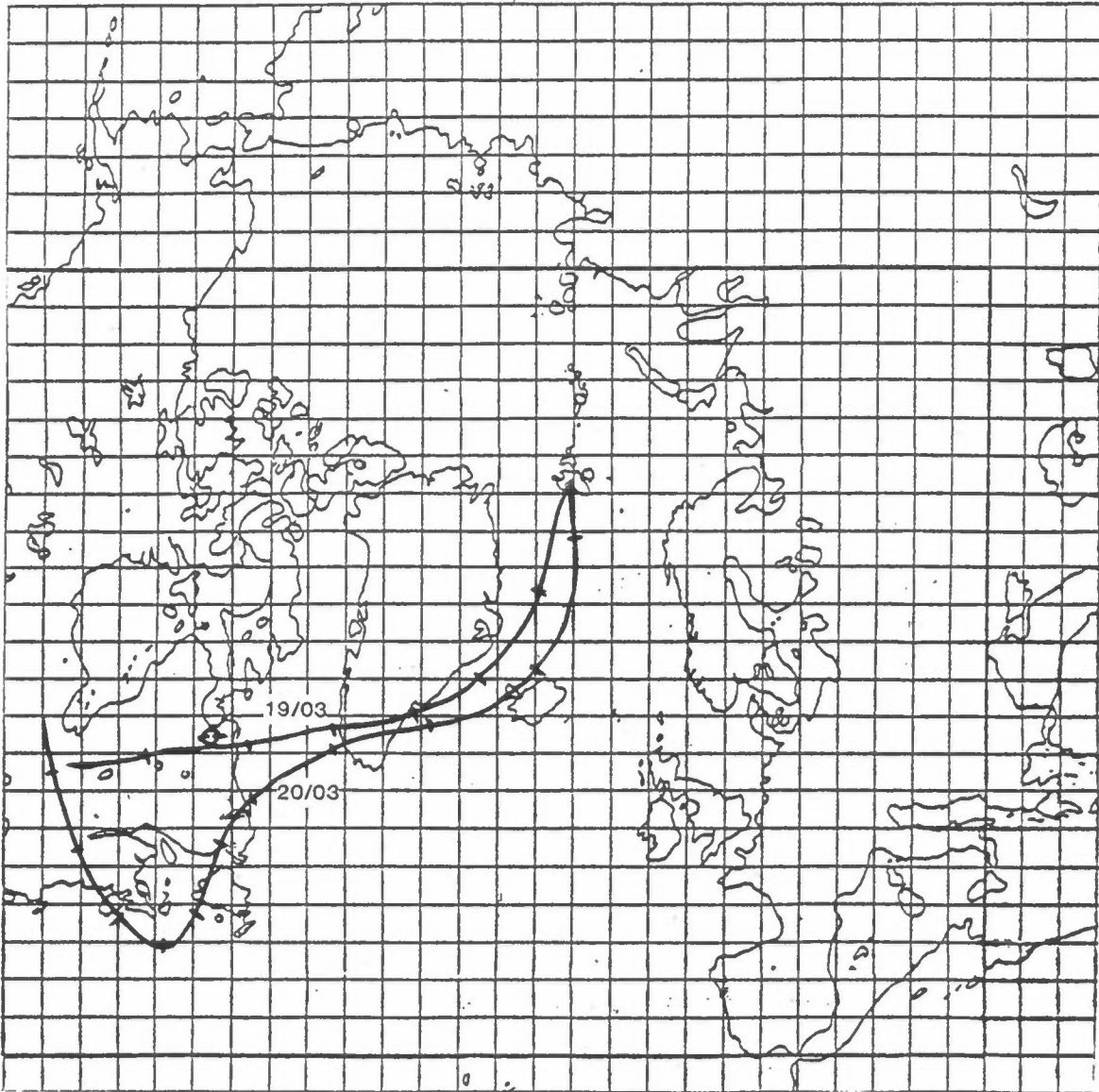


Figure 35: Daily 850 mb trajectories to NYA during the period 1984-03-19 to 1984-03-20.

March 5-6 at KAR in 1984. The above periods fit the time when the increased concentrations of several trace elements were observed at these stations (Pacyna et al., 1985a) due to transport from Western Europe.

The hydrocarbon concentrations peaked again for all mainland stations during March 12-15, 1984. The weather maps indicated air mass passage over Central Europe to Scandinavia. Sharp concentration peaks for anthropogenic trace elements and ions were also observed at JER, KAR and BIR. The hydrocarbon concentrations during this period were similar at all the above stations suggesting low atmospheric deposition "en route".

5.5 JUNE/JULY 1985 CAMPAIGNS

The two- and three-day mean concentrations from this campaign are summarized in the time series in Figures 22-26 for NYA, BJO, JER, KAR and BIR, respectively.

For most of the time in June, the enhanced concentrations of hydrocarbons at BIR and KAR are similar and likely due to emissions from the same source region. The γ - to α -HCH ratios of ca. 1:3 support this finding. The extremely high concentration of fluoranthene at KAR on June 25-26, 1984 is probably a measurement artifact, since no other pollutant showed a similar behaviour. Concentrations of α -HCH at JER are slightly lower, however, two concentration peaks are evident for the period June 18-19 and June 25-26, 1984. Weather maps for the period from the beginning of the campaign until June 21 indicate that high concentrations at that time were caused by anticyclonic circulation over the source areas in Central Europe. From June 22 to June 24 a low pressure centre was located over the Norwegian Sea, directing emissions from the Kola Peninsula in the Soviet Union towards mainland stations of the BP Project.

Enhanced concentrations of α -HCH and 5-PCB at NYA were measured during the period June 20-24 when a low pressure centre was located over Spitsbergen, resulting in a pollution transport from sources in northern Soviet Union. The 850 mb trajectories for NYA are shown in Figure 36. The γ - to α -HCH ratio was ca. 1:17.

1984-06-20 to 1984-06-24

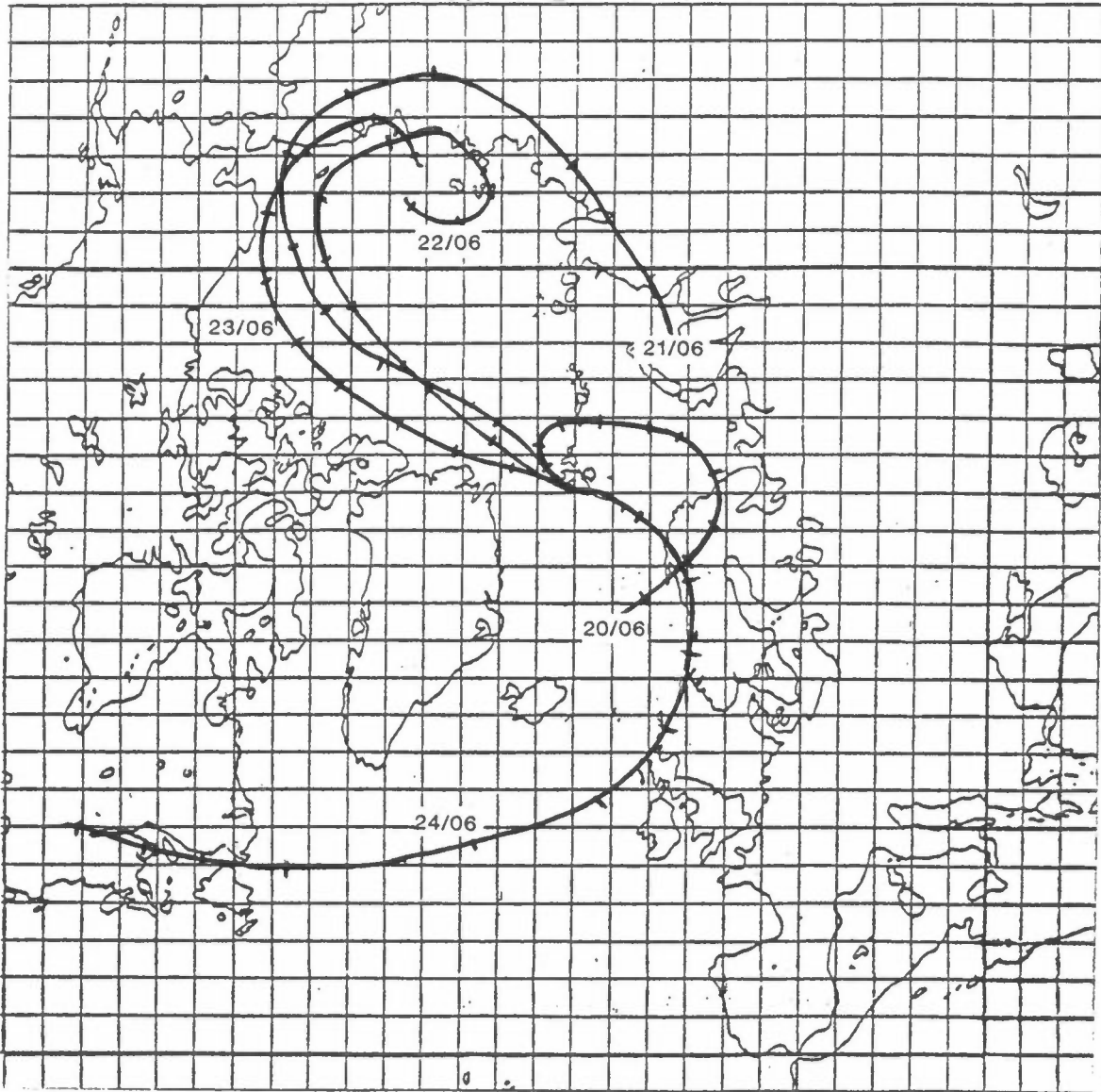


Figure 36: Daily 850 mb trajectories to NYA during the period 1984-05-20 to 1984-06-24.

850 mb trajectories for BJO in Figure 37 indicate transport of air from North America. It is interesting to observe, that the γ - to α -HCH ratio for the air masses from North America was similar for the winter and summer seasons (see section 5.4).

During the period July 7-12, 1984 enhanced concentrations of hydrocarbons were measured at BIR and KAR. At the beginning of the period, anticyclonic circulation dominated Central Europe. Later, between July 9 and July 10, a low pressure centre was situated over the area west of the British Isles. Thus, the increased pollution loads, measured at BIR and KAR were probably due to emissions from various source areas in Europe. This hypothesis is in an agreement with the findings from the BP studies on the origin of inorganic pollutants (Pacyna et al., 1985a). It should be noted that for the first time in our 5 measurement campaigns, the γ -HCH concentrations were higher from the α -HCH concentrations (at BIR July 11-12). The γ -HCH was ranging from 1:4 to 1:2 for KAR and was about 1.0 at BIR. Generally, the α -HCH concentrations were higher at KAR, and γ -HCH at BIR, suggesting a rather fast isomerization.

The high concentrations of hydrocarbons at BIR and KAR were traced at JER on July 11-12. The γ - to α -HCH ratio of 1:4 suggests further isomerization, however, the α -HCH concentration at JER was ca. 2 times lower than that at KAR, indicating "en route" deposition of α -HCH.

Slightly enhanced concentrations of α -HCH, HCB, 5-PCB, α -chlordane and quinones were observed at BJO during the period July 9-12. The 850 mb trajectories to BJO at that time, shown in Figure 38, indicate transport of the arctic air to the island.

1984-06-22 to 1984-06-26

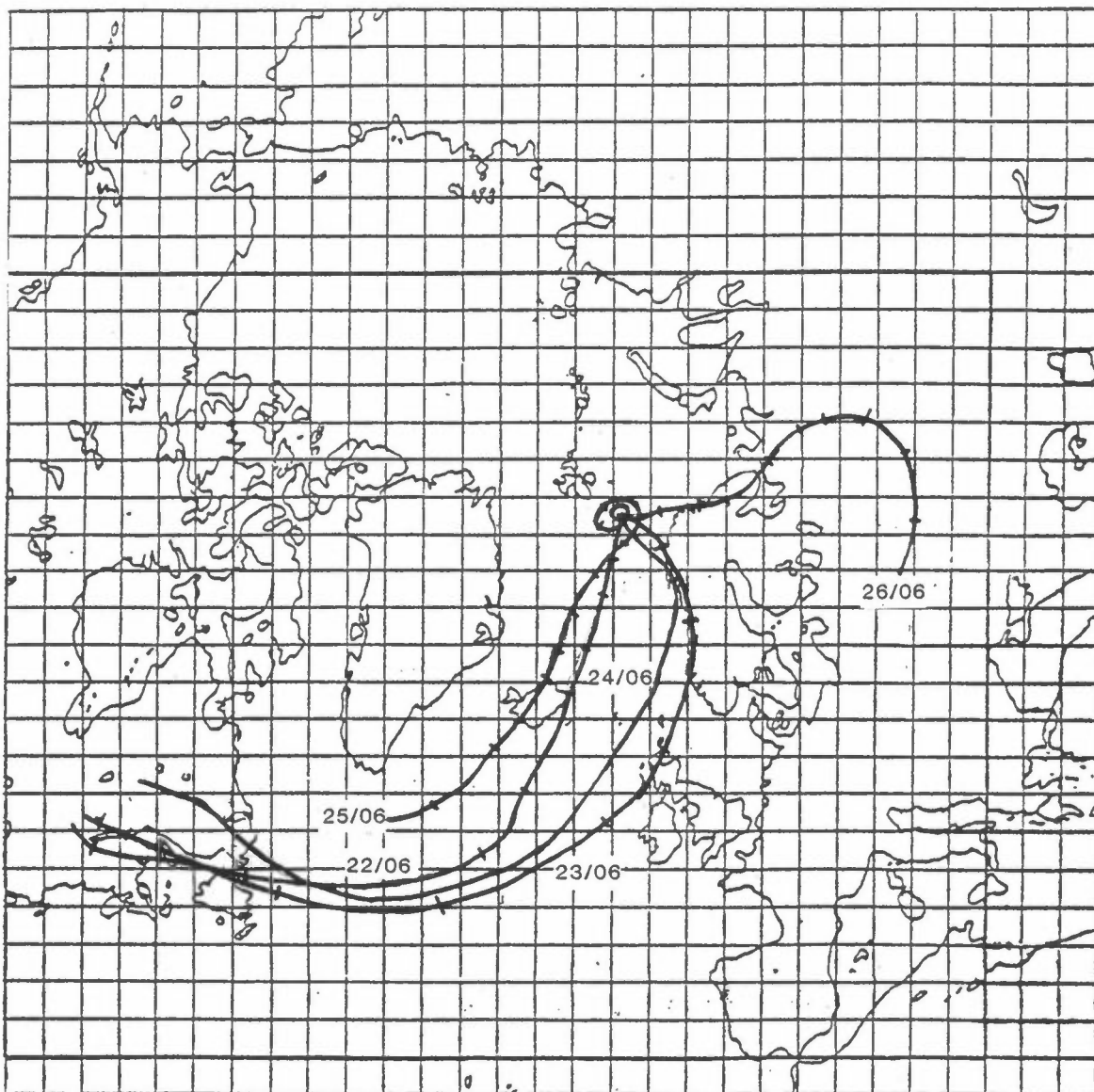


Figure 37: Daily 850 mb trajectories to BJO during the period 1984-06-22 to 1984-06-26.

1984-07-09 to 1984-07-12

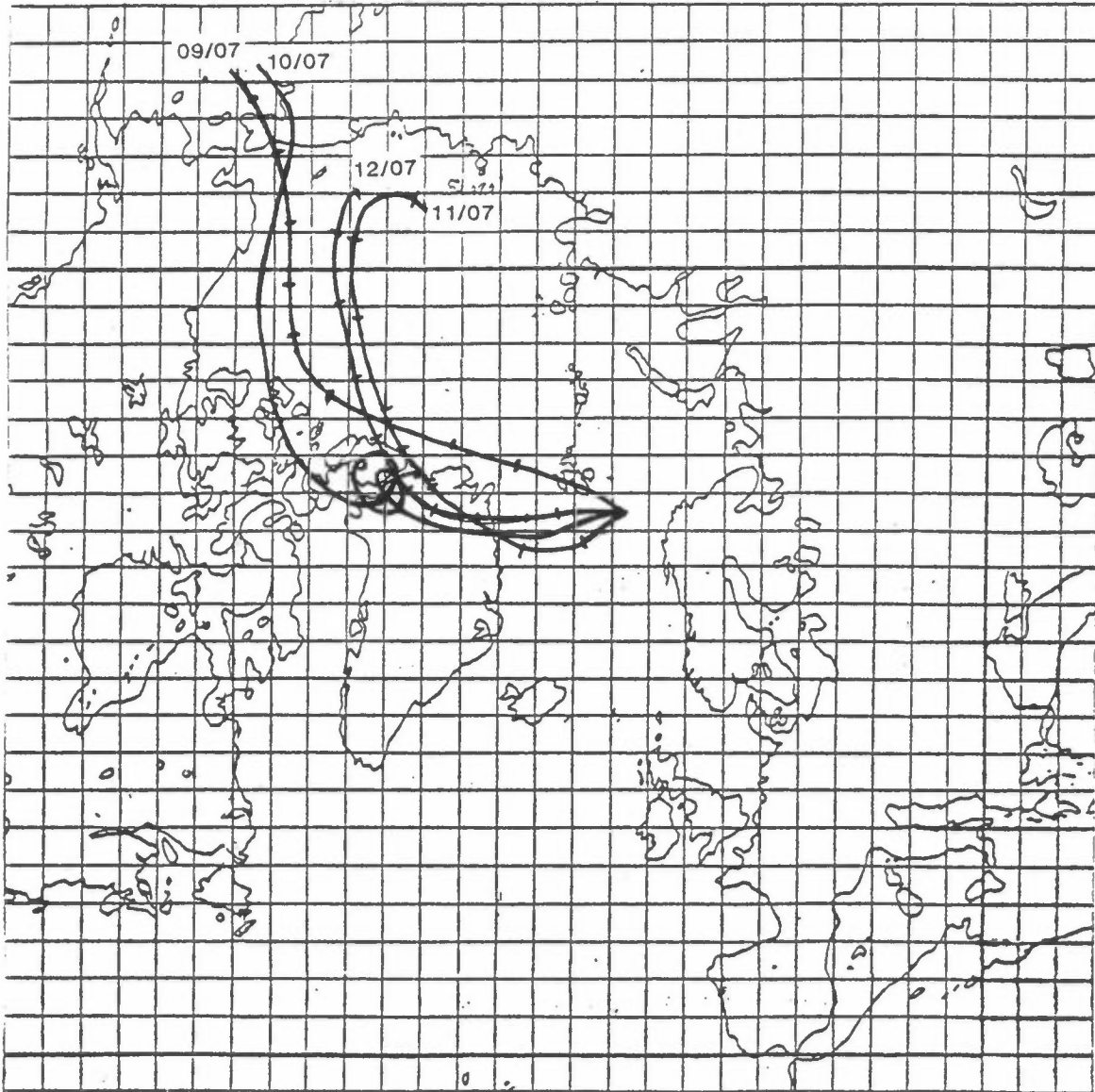


Figure 38: Daily 850 mb trajectories to BJO during the period 1984-07-09 to 1984-07-12.

6 SEASONAL VARIATIONS OF HYDROCARBON CONCENTRATIONS

Mean concentrations and concentration ranges measured during the BP Project campaigns at NYA and BJO are shown in Tables 2 and 3, respectively. It is interesting to note the very significant decrease of the α -HCH concentrations at both stations and the increase of the γ -HCH concentrations after Fall 1982. Since most of the air pollution at NYA and BJO during the summer-fall season is related to emissions in Europe (Pacyna et al., 1985a), it can be suggested that changes in the α - and γ -HCH concentrations at both stations are due to the use of more purified HCH as insecticide in Europe. Thus, less α -HCH isomer seems to be mixed with more γ -HCH in order to produce and apply pesticides in the European countries after 1982. However, this suggestion is not confirmed due to the lack of statistical information.

The mean concentrations in Tables 2 and 3 are dependent upon episodic concentrations measured during our campaigns. The strongest episode was observed during the period March 6-13 in 1983 when the air masses were carrying pollutants emitted from sources in the northern Soviet Union (Pacyna et al., 1985a). This explains the highest concentrations of γ -HCH, 5-PCB, fluoranthene and quinones at NYA and BJO in Winter/spring 1983.

The concentrations of fluoranthene and quinones are higher for the winter/spring than in the summer/fall season at both stations due to emissions from combustion processes from local and remote sources. Only small changes are found in the concentrations of HCB, α -chlordane and 5-PCB, except the 5-PCB values measured in winter/spring 1983, explained earlier in this section. This indicates that the existence of HCB, α -chlordane and 5-PCB in the arctic air is more due to general influx of air masses to the Arctic than governed by episodic transport.

The concentration ratio of γ - to α -HCH seems to be a useful tool to distinguish between sources of air pollutants measured in the Arctic. Ratios of γ - to α -HCH ranging from 1:4 to 1:1 were found for the "European" air pollutants, 1:10 to 1:7 for North America and 1:10 to less than 1:100 for the Soviet Union. These ratios were higher in the winter/spring than in the summer/fall seasons indicating that isomerization of γ -HCH to α -HCH is more efficient in the summer/fall season.

Table 2: Concentration ranges and mean values measured at Ny Alesund during 5 measurement campaigns (in $\text{pg} \cdot \text{m}^{-3}$).

Campaign	α -HCH		γ -HCH		HCB		5-PCB		α -chlordane		Fluoranthene		Quinones	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Fall 1982	757 - 1416	1192	0.1 - 9.4	2.1	75 - 169	123	2.3 - 46.5	15	0.6 - 3.2	1.2	8.2 - 134	43	73.5 - 364	217
Fall 1983	407 - 695	548	34 - 67	46	158 - 227	186								
Summer 1984	260 - 774	488	24 - 100	47	20 - 201	154	0.9 - 43.0	21	1.7 - 5.4	2.8	0.4 - 46	20	52 - 610	217
Winter/ Spring 1983	151 - 734	485	21 - 102	66	112 - 187	144	32 - 145	62	1.1 - 2.3	1.7	13 - 145	57	38 - 952	325
Winter/ Spring 1984	121 - 787	273	12 - 70	29	29 - 389	151	4 - 51	16	0.6 - 5.1	2.0	12 - 142	48	21 - 1040	267

Table 3: Concentration ranges and mean values measured at Bjørnøya during 4 campaigns (in $\text{pg} \cdot \text{m}^{-3}$).

Campaign	α -HCH		γ -HCH		HCB		5-PCB		α -chlordane		Fluoranthene		Quinones	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
Fall 1982	420 - 1550	774	0.1 - 29	4.2	78.4 - 200	120	6.3 - 37.6	17	0.5 - 1.7	0.8	17.6 - 143	47	66.6 - 236	145
Fall 1983	277 - 477	326	14 - 32	22	88 - 180	134								
Summer 1984	38 - 305	155	5 - 41	18	42 - 149	57	4.4 - 14	8	0.6 - 2.1	1.3	12 - 43	17	68 - 458	185
Winter/ Spring 1983	110 - 469	282	23 - 80	47	87 - 201	124	8.5 - 66	23	1.2 - 3.1	1.7	42 - 359	139	107 - 1646	590

7 GEOGRAPHICAL VARIATIONS OF HYDROCARBON CONCENTRATIONS

The hydrocarbon concentrations at NYA and BJO in Tables 2 and 3 can be compared with the concentrations measured in other parts of the world. Some data for α - and γ -HCH and HCB in the urban air, the air over the areas with enhanced use of pesticides and in clean air (Antarctica) are presented in Table 4. All these concentrations were measured before the BP measurement campaign in Fall 1982, and represent mean values for the winter and summer seasons.

The α -HCH concentrations in the Arctic are at the same level as concentrations measured in the U.S.A. The concentrations in Lillestrøm, Southern Norway, were measured in May 1981 and are in the range observed at NYA and BJO in Fall 1982. However, the γ - to α -HCH ratio is 1:3 for Lillestrøm, indicating a contribution of lindane from European or local sources.

Concentration ratios ranging from 1:17 to 1:8, were observed in the air over the Northwest Pacific and the western coast of India (Tanabe et al., 1982), suggesting the use of pesticides of a composition different from that in Europe.

The concentrations measured in the air over the Antarctic Ocean are low and it is difficult to speculate on their origin.

The HCB concentrations in Tables 2, 3 and 4 are very similar indicating a very even distribution of HCB over the world. There is not only similarity in the geographical distribution of the HCB concentrations but in seasonal variations as well.

Table 4: Concentrations of α - and γ -HCH and HCB in various places of the world (in pg.m^{-3})

Location	α -HCH	γ -HCH	HCB
1. USA			
Denver, CO (Billings and Bidleman, 1983)	300 ^{*1}		240
New Bedford, MA (Billings and Bidleman, 1983)	1000 ^{*1}		180
Columbia SC (Bidleman, 1981)	370	180	200
College Station, TX (Atlas and Giam, 1981)	420		200
2. NORWAY			
Lillestrøm (Oehme and Ottar, 1984)	875	318	162
3. NORTHERN PACIFIC OCEAN			
- Enewetak Atoll (Atlas and Giam, 1981)	250	15	100
4. WESTERN PACIFIC OCEAN			
(Tanabe et al., 1982)	1000	100	
5. EASTERN INDIAN OCEAN			
(Tanabe et al., 1982)	500	64	
6. ANTARCTIC OCEAN			
(Tanabe et al., 1982)	25	9	
7. NORTH ATLANTIC			
(Atlas and Giam, 1981)	390		150

*1 α and γ -HCH

8 CONCLUSIONS

Several instances of concentration peaks for hydrocarbons were observed during the various BP Project campaigns, indicating either long-range transport of air pollutants or emissions from local sources. In the case of α - and γ -HCH, HCB, 5-PCB and α -chlordane, their enhanced concentrations observed sporadically at the arctic island and at the Norway mainland stations, were due to long-range transport of air masses over regions with intense use of pesticides, such as Asia, Europe and North America. In order to assess the origin(s) of the air pollutants measured during these episodes, the concentration ratio of γ - to α -HCH was examined, together with relevant information from weather maps and/or wind back-trajectories for these periods. Emissions from the Soviet Union sources seem to be a major contributor to the episodic concentrations of hydrocarbons at the arctic island stations during both wintertime and summertime. Sources in Europe can be assigned as main contributors to the concentrations observed at the Norway mainland stations in the summer half-year. Traces of emissions from the North American sources are sporadically found in the Norwegian Arctic.

Generally more than 98% of all compounds were found in the vapour phase. The HCH concentrations were higher in summertime than in wintertime due to the use of pesticides in the spring season.

The high concentrations of fluoranthene and quinones observed on some occasions, particularly during wintertime, are related to the emissions from combustion processes in the Svalbard Archipelago and in more remote regions, e.g. northern Soviet Union. A good correlation between concentrations of fluoranthene and quinones at Hopen is possibly due to the occurrence of land-slides caused by the permafrost ground.

The Arctic seems to be an area where hydrocarbon deposition takes place. The atmospheric residence time of the measured hydrocarbons is long enough to explain their transport from very far away sources and slow deposition "en route" and in the Arctic. In summer, the concentrations of the more volatile compounds seem to be significantly influenced by sublimation of previously deposited materials.

It is difficult to conclude on the γ - to α -HCH isomerization during their long-range transport to the Arctic. There is, however, evidence from the data in this report that such a process does exist. The rate of isomerization is faster in summer than in winter. The half-life of the γ -HCH may be as short as a few days in summer and probably no isomerization takes place in the arctic winter. Generally, our knowledge of the γ - to α -HCH isomerization in the air is very incomplete.

These conclusions are in good agreement with the findings reported from other BP Project studies, e.g. on the origin of aerosols aloft (Pacyna et al., 1985b), chemical composition of aerosols at ground stations (Pacyna et al., 1985a) and statistical analysis of aerosol sulphate in the Norwegian Arctic and long-scale atmospheric flows (Iversen, 1985).

9 ACKNOWLEDGEMENTS

This work was financed by British Petroleum International Ltd.

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NORSK INSTITUTT FOR LUFTFORSKNING (NILU)
 NORWEGIAN INSTITUTE FOR AIR RESEARCH
 POSTBOKS 130, 2001 LILLESTRØM (ELVEGT. 52), NORGE

RAPPORTTYPE Oppdragsrapport	RAPPORTNR. OR 19/86	ISBN-82-7247-685-1	
DATO March 1986	ANSV. SIGN. <i>J. Schjordegen</i>	ANT. SIDER 77	PRIS NOK 60,00
TITTEL Organic air pollutants in the Norwegian Arctic.		PROSJEKTLEDER M. Oehme	
		NILU PROSJEKT NR. O-8512	
FORFATTER(E) J. M. Pacyna M. Oehme B. Ottar		TILGJENGELIGHET* A	
		OPPDRAGSGIVERS REF.	
OPPDRAGSGIVER (NAVN OG ADRESSE) BP International Ltd., ECC Britannia House, Moore Lane London, EC24 9BV, England			
3 STIKKORD (à maks. 20 anslag) Arctic Air quality measurement Pesticide			
REFERAT (maks. 300 anslag, 7 linjer)			

TITLE
<p>ABSTRACT. Atmospheric emission sources and transport within air masses of some polychlorinated hydrocarbons, fluoranthene and quinones are reviewed and their importance for the Arctic is discussed. The origin of hydrocarbons in the Arctic is examined with the help of measured concentration ratios of γ- to α-HCH during 5 measurement campaigns in Fall 1982 - Summer 1984 at stations in the Arctic and the Norwegian mainland, and information from weather maps and wind back-trajectories.</p>

*Kategorier: Apen - kan bestilles fra NILU A
 Må bestilles gjennom oppdragsgiver B
 Kan ikke utleveres C