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Organic compounds in rainfall at Hornsund, SW Spitsbergen: qualitative results

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Abstract: Research on the chemistry of atmospheric precipitation in the Hornsund region of Svalbard has been extended by analysis of the organic contents. In rainfall samples collected in September 2003, the organics were separated by solid phase extraction (SPE), eluted and analysed on gas chromatograph coupled to a mass selective detector (GC/MS). Rainfall pH was in the range 4.72–5.45, the low values suggesting possible pollution. Concentrations of inorganic ions, expressed as total dissolved salts (TDS), were 5.40–13.18 mg L^{-1}. Non-sea-salt (nss) sulphates were in the range 5–11 μeq L^{-1}. In all samples, long-chain alkanes with chain length up to C_{36}, and their methyl derivatives were detected. Among aromatic compounds biphenyl, dibenzofuran and its methyl derivatives were found. Polycyclic aromatic hydrocarbons (PAHs) were represented by naphthalene, phenanthrene, fluorene, acenaphthene, fluoranthene and pyrene. There were no PAHs with higher numbers of rings. The synoptic meteorological conditions in September 2003 indicate that all organic and inorganic pollutants were of local origin.

Key words: Arctic, Svalbard, Hornsund, organic compounds, rainfall, chemistry.

Introduction

Research on organic compounds in Svalbard focuses chiefly on their presence in animals. Levels of polychlorinated biphenyls (PCBs) and DDT have been determined in polar bears (Derocher *et al.* 2003, Haave *et al.* 2003, Lie *et al.* 2003), birds (Borgå *et al.* 2003, Herzke *et al.* 2003) and seals (Sørmo *et al.* 2003). Less attention is given to Polycyclic Aromatic Hydrocarbons (PAHs) because these compounds have not been accumulating for a long time in living tissues and, in any case, undergo bacterial decomposition. However, the potential of PAHs to form carcinogenic and mutagenic diols and epoxides that react with DNA (Macdonald *et al.* 2000) demands a greater knowledge of its concentrations in the Arctic environment. Natural sources of PAHs include volcanoes, burning of biomass, losses or seeps of petroleum or coal deposits (Macdonald *et al*. 2000). These compounds

can also be results of diagenesis of soil and sediments (Masclet *et al.* 2000). PAHs in the atmosphere derive primarily from incomplete combustion of fossil fuels to produce electricity and heat, vehicular exhausts, industrial activities and forest fires.

PAHs dominant in the Arctic are found in the air (Patton *et al.* 1991; Halsall *et al.* 1997; Macdonald *et al.* 2000), snow (Peters *et al.* 1995; Welch *et al.* 1991; Masclet *et al.* 2000) and ice (Kawamura *et al.* 1994; Masclet and Hoyau 1994; Isaksson *et al.* 2003). Two- and three-ring PAHs are usually microbiologically decomposed (AMAP 2004). PAHs of high molecular weight tend to be attached to atmospheric particles. These less volatile PAHs belong to one-hop contaminants but many PAHs behave as multi-hop contaminants, returning to the atmosphere after initial deposition (Macdonald *et al.* 2000).

On Svalbard, local coal mining activities and settlements are the major primary sources of PAHs, with coal-fired power plants being located in five settlements (AMAP 2004). However, depending on the synoptic meteorological conditions, PAHs detected in the Arctic air may also reflect contributions of emissions from middle and high latitudes (Halsall *et al.* 1997; Masclet *et al.* 2000).

The Lomonosovfonna 122-meter ice core, representing ice which has accumulated in the last century, contained naphthalene in concentrations ranging 5–53 ng kg^{-1}, *i.e.* about fifty times higher than in Greenland (Vehviläinen *et al.* 2002). Naphthalene data showed very low concentrations up to the mid-1930s, higher values around 1980 and a possible decrease in recent times (Vehviläinen *et al.* 2002).

Fluoranthene was detected in air sampled at Ny-Ålesund in the winter/spring of 1983 and 1984 (Pacyna and Oehme 1988). PAHs in the air are measured weekly at Zeppelin Station in Ny-Ålesund; around 39 compounds from this group have been detected, plus PCBs and 13 chlorinated pesticides. Naphthalene and biphenyl represent approximately 50% of the sum of PAHs. Results (presented on *www.nilu.no/niluweb/services/zeppelin*) show a decrease of the yearly average of the sum of PAHs from 6.4 ng m^{-3} in 1997 to 3.1 ng m^{-3} in 2001. This is accompanied by a decrease in the SO_4^{2-} concentration in atmospheric precipitation collected in Ny-Ålesund. Higher concentrations of PAHs in the air were found in winter.

Seasonal variations in the abundance of pollutants in Svalbard air occur because the lower tropospheric circulation in the Arctic in winter differs from that in summer. During the winter, it is dominated by high pressure over the continents and lows over the oceans. As a result, an atmospheric conduit is formed to transport polluted air masses from Siberia and Eastern Europe to Svalbard (AMAP 2004). The Polar Front does not limit transport of pollutants into the Arctic. During the summer, because the oceanic low-pressure cells weaken and the continental high-pressure cells disappear, northward transport from low latitudes weakens. The Polar Front is then situated further north. This creates a meteorological barrier

which is difficult for air masses from the southern, polluted regions to penetrate (AMAP 2004). Photochemical degradation occurring during the 24-hour summer daylight period may be another reason for the much lower concentration of PAHs.

The first comprehensive chemical analyses of summer rainfall in Hornsund were made in 2000, and have been discussed in relation to atmospheric circulation patterns by Krawczyk *et al.* (2002) and Głowacki and Krawczyk (2002). Inorganic pollutants were found (*e.g.* non-sea-salt (nss) sulphate) and, after a full year of sampling, it was decided to expand this research to organic compounds in rainfall because we haven't found such published data from the Arctic. According to Macdonald *et al.* (2000) measurements of contaminants in the rain for PAHs and organochlorine compounds have not been made in the Canadian Arctic.

Methods

Five rainfall samples with volumes of 2–3 L each were collected, during the storms that occurred in 2003, on September 4, 8, 9, 10 and 11. The samples were taken in glass bottles via a polypropylene funnel with an area of 0.25 m^2. The sampling site was situated in the Fuglebekken catchment, about 500 m NE of the Polish Polar Station in Hornsund (77°0.386'N, 15°33.178'E).

One sample of surface water was taken on July 16, 2003, from the Fuglebekken creek at the main hydrometric site in the catchment. This sample was collected into two PET bottles with a total volume of 3 L and, after a few hours, transferred into three 1 L glass containers.

Part of this sample was used for measurement of pH, SpC and inorganic ions analysis. Samples were passed via 0.45 μm mixed cellulose ester membrane filters (Advantec MFS, USA) and divided into two aliquots. One aliquot was acidified with HNO_3 for cation analysis, carried out with an ion chromatograph Compact IC 761 (Metrohm, Switzerland), Cation 1–2 column, tartaric acid/dipicolinic acid eluent. Anions were analysed in untreated samples, using the Metrohm Compact IC 761 in suppressor mode, an ASUPP5 column, $NaHCO_3/Na_2CO_3$ eluent.

Organic compounds were separated from the aqueous phase by the solid phase extraction (SPE) method, using Bakerbond C18 columns (3 g octadecyl on silica gel carrier) purchased from J.T. Baker (S.Witko – Łódź, Poland). The Bakerbond Application Note EN-19 (Extraction of PAHs from drinking water) was used for extraction procedures. Total recovery of PAHs >85% is assumed for these columns. The rainfall samples were passed through at a rate 8–10 cm^3 per minute. Organic compounds were eluted from the column with several portions of dichloromethane (~0.5–1 cm^3), and their eluates were combined. To ensure complete elution of the organics the SPE columns were checked under UV light for fluorescence of aromatic hydrocarbons. The solvents from the eluates were evaporated at room temperature. The organics were analysed by GC/MS on an Agilent Technol-

ogy 6890 gas chromatograph coupled with Agilent 5973 network mass sensitive detector. The HP-5 column (60 m × 0.25 mm), coated by a 0.25 μm stationary phase film was used. All solvents used were analar or specpure grade (POCh, Poland). Water was prepared by a Milli Q/RO type device, made by Millipore Ltd. (USA). Carrier gas used was helium 6.0 from Linde Gas Poland, with O_2, N_2, $H_2O \leq 0.5$ ppm and C_nH_m, CO, $CO_2 \leq 0.1$ ppm. Temperature program started at 50˚C kept for 2 min, then raised to 175˚C – at 10˚C min^{-1}, then to 225˚C – at 6˚C min^{-1}, and then to 300˚C at 4˚C min^{-1}, final isothermal temperature 300˚C was held for 20 min. The mass spectrometer was operated in the electron impact ionisation mode at 70 eV and scanned the mass range from 50 to 650 da. Data were acquired in a full scan mode and processed with the Hewlett Packard Chemstation software. The SCAN mode was selected because the purpose of this preliminary and qualitative research was to check if any PAHs will be detected in Hornsund rainfall. All compounds were identified by their mass spectra, by comparison of retention times of their peaks to those of standard compounds and to the literature, and by interpretation of MS fragmentation patterns (Wiley 2000). The contact of rainfall with polypropylene funnel was short but has given mass spectra of eluted compounds. Retention times of these compounds (mostly phtalates) were known from earlier research and this "background" was removed.

The sensitivity performance was verified with 1 pg μL^{-1} solution of octafluoronaphthalene (OFN).

Results

Meteorology. — A trend to increasing average monthly temperature for September is being observed at Hornsund, together with some increase in summer precipitation totals (Głowacki and Krawczyk 2002). In August 2003, the average monthly air temperature in Hornsund was 4.3˚C, whereas in September it had reduced to 1.2˚C (Table 1). August was unusually dry, with precipitation totaling only 32.7 mm, and none recorded after August 14 (Fig. 1). There was substantial rainfall during the first part of September, succeeded by 10 days without precipitation following September 17 (Fig. 1). On September 27, the first snowfall was recorded, which turned into 7.4 mm of rain on September 28 and 29. Monthly precipitation in September 2003 totaled 66.0 mm.

Table 1

Mean monthly air temperatures and precipitation totals in Hornsund, Svalbard in August and September, 2003. Data from the Polish Polar Station in Hornsund.

	T_{mean} [˚C]	T_{max} [˚C]	T_{min} [˚C]	P [mm]
August	4.3	6.3	2.7	32.7
September	1.2	2.7	–0.6	66.0

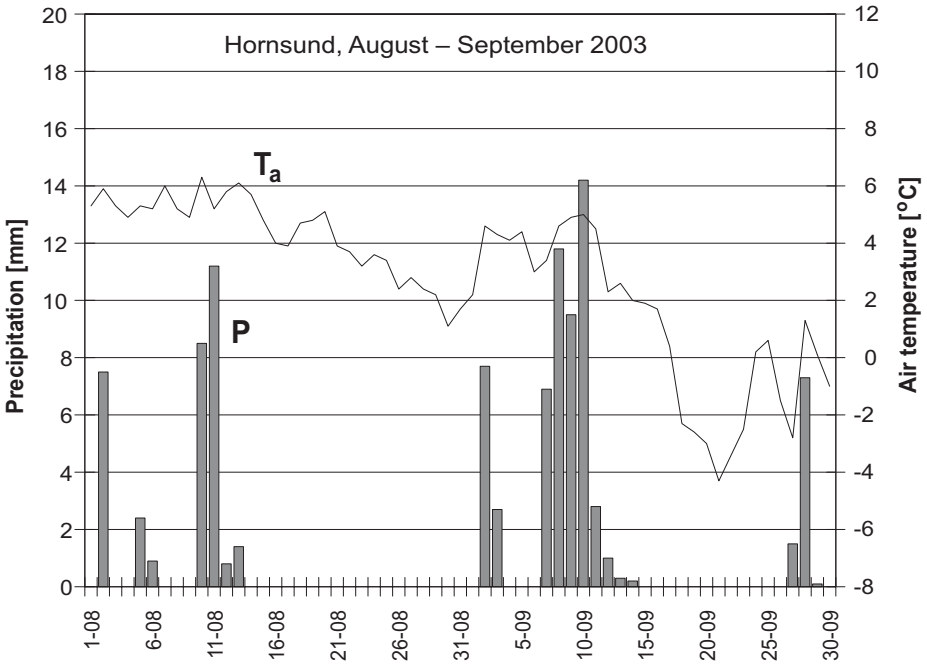

Fig. 1. Plot of mean daily air temperature and precipitation totals in Hornsund in August and September 2003. Data from the Polish Polar Station in Hornsund.

Synoptic maps for the North Atlantic (www.wetterzentrale.de) show that on September 3 and 4 low pressure dominated over Svalbard and the atmospheric circulation was only local, with air masses moving around the archipelago. On September 7 and 8, there was a strong front south of Svalbard, which retarded movement of air masses from the South, thereby permitting a local SE to NW circulation over the archipelago. On September 9, this atmospheric front moved towards the North Pole, allowing SW to NE air flow from the Greenland Sea. On September 10, the high pressure center had formed South of Svalbard and air flow direction was from W to E. On September 11, two fronts developed over the archipelago. The highest rainfall was on September 10 (14.2 mm), the least (1.7 mm) on September 4 (Fig. 2).

Inorganic compounds in rainfall. — Rainfall pH ranged 4.72–5.45, with a mean volume weighted (v/w) value of 4.89, somewhat higher than the v/w pH = 4.70 recorded in the summer of 2000. Concentration of H^+ was in the range, 3.5–19.1 μeq L^{-1}, and specific conductivity (SpC), 8.90–23.35 μS cm^{-1}, (mean v/w SpC = 15.28 μS cm^{-1}). In all the September rainfall events, Na^+ and Cl^- were the dominant ions, as is perhaps to be for a station located on the edge of a fjord. Comparison with mean (v/w) concentrations of ions in 18 rainfall events from the summer of 2000 and in 12 rainfalls of 2001 (Table 2) shows similar ranges. Summer 2000 concentrations of marine salts were higher, most probably because of very

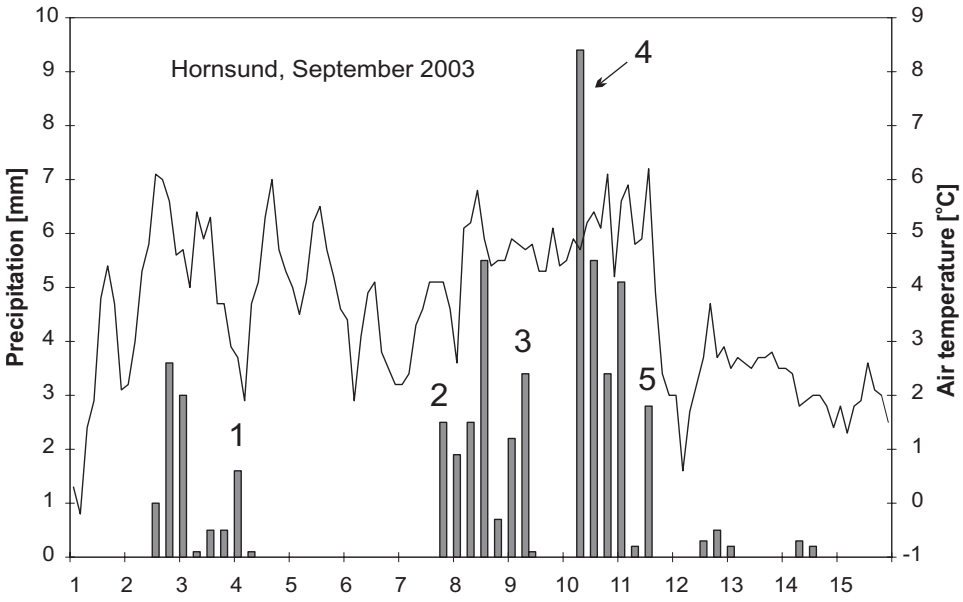

Fig. 2. Rainfall events sampled for analysis on the background of air temperature and precipitation totals, September 2003. Data from the Polish Polar Station in Hornsund.

Table 2

Chemical characteristics of rainfall in Hornsund in September 2003: amount of rainfall (in mm), pH, ionic concentrations (in μeq L^{-1}) - nss-SO_4^{2-} concentration is given in parentheses, total dissolved salts (TDS) concentration (in μg L^{-1}). For comparison average (v/w) concentrations in the summers of 2000 (n = 18) and 2001 (n = 12). Data for 2000 and 2001 from Głowacki and Krawczyk (2002).

	P	pH	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	NH_4^{+}	HCO_3^{-}	SO_4^{2-}	Cl^{-}	NO_3^{-}	TDS
September 4	11.4	5.45	17	25	76	5	19	50	14 (6)	82	2	934
September 8	14.5	4.96	18	47	132	2	0	65	22 (11)	110	4	1318
September 9	7.0	4.81	13	15	42	1	0	30	10 (5)	44	1	540
September 10	18.6	4.72	4	13	59	2	0	20	13 (7)	64	3	594
September 11	8.6	4.92	6	15	70	2	0	10	15 (7)	74	3	610
Mean 2000		4.70	12	26	117	4	nd	–	21	134	1	
Mean 2001			10	6	40	2	nd	–	11	44	6	

windy conditions that year. In September 2003 salts of marine origin constituted 52% (September 9) to 77.7% (September 11) of total dissolved salts. Sulphate concentrations in September 2003 rainfall were lower than in the summer of 2000, where long-range transport of atmospheric pollution from Central and Eastern Europe was detected (Krawczyk *et al.* 2002). Nss-sulphates were in the range, 5–11 μEq L^{-1}; no correlation was found with rainfall pH.

Only in the rainfall of September 4, the ammonium ion NH_4^+ was found. It is presumed that this has been generated by bacterial decomposition of bird droppings. Ammonia concentration in the air probably could increased during this three week long dry period in August, resulting in the presence of NH_4^+ in the first rainfall.

Organic compounds in rainfall. — The greatest number of different organic compounds, both aliphatic and aromatic, extracted on C_{18} phase, was found in the rainfalls of September 4 and 10 (Table 3). The lowest number of organic compounds was extracted from the September 9 rains at a time when air masses were moving from the region of the Greenland Sea.

The first sample (September 4) rich in organic compounds, was taken during the first rainfall following a twenty-day dry period. The rainfall containing only alkanes and phenanthrene has fallen in the middle of rainy period on September 9, when 13.5 mm of rain has already scavenged the atmosphere.

In all rainfall samples, long-chain n-alkanes and its methyl derivatives were found (Table 3), with chains up to as much as C_{36} (September 9). An example of their distribution is shown in Fig. 3. N-alkenes with one double bond were found only in September 10 rain. In the rainfall sample of September 4, a series of fatty acids containing palmitic (C_{16}), stearic (C_{18}) and oleic (C_{18}) acids were identified. These are believed to probably derive from pollens of tundra plants. Biphenyl was

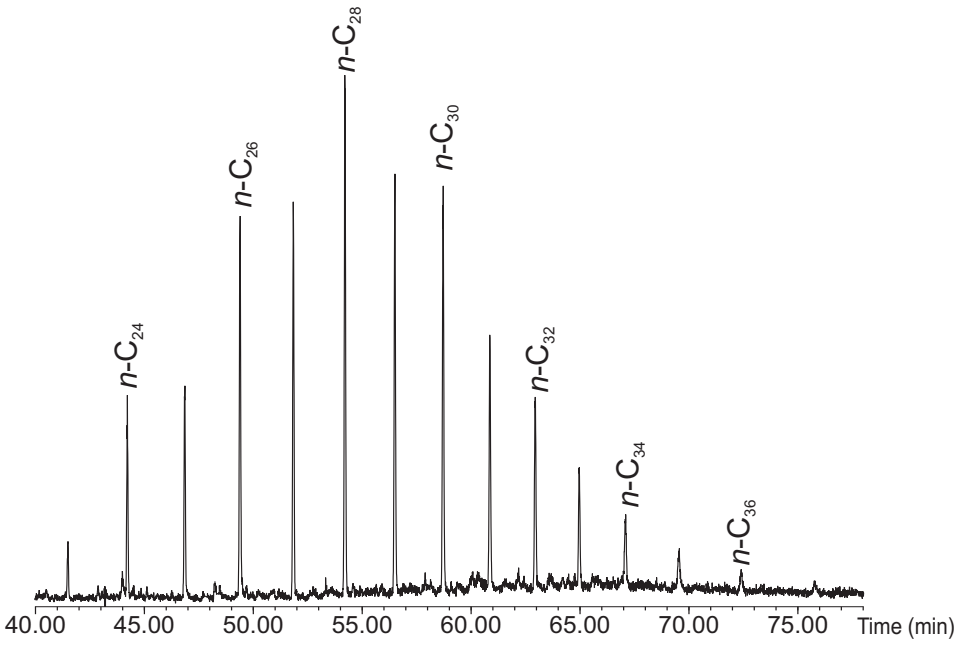

Fig. 3. Example of distribution of n-alkanes found in rainfall at Hornsund on September 8, 2003.

Table 3

Organic compounds found in Hornsund rainfall, September 2003.

	September 4	September 8	September 9	September 10	September 11
n-alkanes	+	+	+	+	+
n-alkenes				+	
Fatty acids	+				
Biphenyl	+	+		+	+
Dibenzofuran	+	+			+
Naphthalene	+	+		+	
Phenanthrene	+	+	+	+	+
Fluorene	+			+	+
Acenaphthene	+			+	
Pyrene				+	
Dibenzothiophene		+			

not detected in the rainfall of September 9. Biphenyl and dibenzofuran were accompanied by its methyl derivatives (Fig. 4).

Among PAHs, only phenanthrene was found in all of the rainfall samples (Table 3), and was always accompanied by its methyl derivatives. Naphthalene and fluorene were detected in three samples, acenaphthene only in two, pyrene in one (September 10), dibenzothiophene also in only one sample (September 8). No PAHs with a number of rings greater than four were found in the samples analysed.

The single analysed sample of surface water from Fuglebekken stream contained organic compounds similar to those in the rainfall. Water flowing through the tundra contained mainly n-alkanes (C_{11}–C_{30}) and their alkyl derivatives, n-alkenes and branched alkanes. Among aromatic compounds, phenanthrene, biphenyl and dibenzofuran with its methyl derivatives were identified.

Concluding remarks

Organic compounds, both aliphatic and aromatic, were found in rainfall samples collected in Hornsund. The highest number of organic compounds was found in rainfalls on September 4 and 10, whereas the sample with the highest inorganic ion concentration (TDS=13.18 mg L^{-1}) and nss-sulphate (11 μeq L^{-1}) was taken on September 8. Long-chain alkanes and phenanthrene were detected in all rainfall samples. PAHs found in Hornsund rainfall are the same as in the air samples in the Canadian Arctic. The annual mean air PAH concentrations (both vapour and particulate phases) for Alert, Tagish and Dunay in 1993 show the highest values for

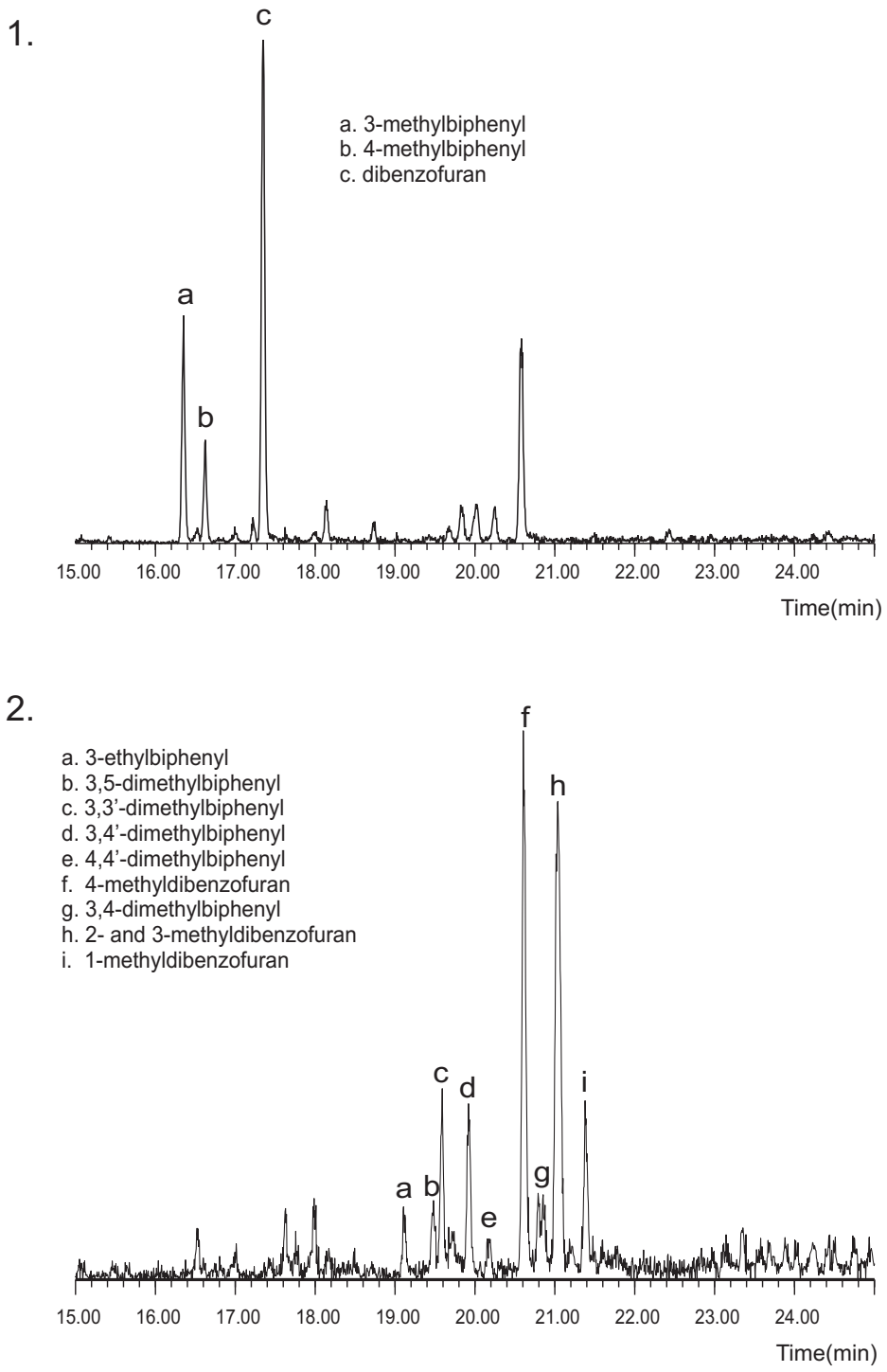

Fig. 4. Example of distribution of aromatic compounds in rainfall at Hornsund on September 11, 2003.

phenenthrene and fluorene; acenaphthene and pyrene have also been present (Macdonald *et al.* 2000).

The synoptic conditions when the rainfall samples were taken in September 2003 indicate that all organic and inorganic pollutants must have been of local origin. The rainfall with the high number of extracted organic compounds was associated with the occurrence of atmospheric lows. The first rainfall sampled had the highest number of organic compounds (September 4), following three weeks of dry weather. The lowest number of organic compounds in the rain, extracted on the C_{18} phase, was found on September 9, when air masses arrived from the Greenland Sea.

We can suppose that the concentrations and number of organic compounds found in rainfall at Hornsund are the lowest ones. Snowfall samples collected during winter should have higher concentrations because snow is a better scavenger and atmospheric circulation during winter favours inflow of pollutants. At Alert in the Canadian Arctic the mean PAHs concentration in air during the cold period was an order of magnitude higher than the warmer season (Macdonald *et al.* 2000).

When estimating the amount of carbon dioxide sequestered from the atmosphere during chemical weathering processes of carbonate and silicate rocks in polar regions, one should bear in mind that a part of this CO_2 can result from oxidation of organic compounds in water. We have shown that organic substances are present in Svalbard precipitation and runoff. Polycyclic aromatic hydrocarbons are detected not only in the ice cores but also in rainfall and surface waters in an area covered by tundra.

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