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Petroleum hydrocarbon concentrations in Baltic Sea subsurface water

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Many oil hydrocarbons are thought to be a source of toxicity in the marine environment. The Baltic Sea, one of the largest bodies of brackish water in the world, has numerous point sources for oil pollution. In particular, growing maritime traffic, including oil transportation, and several oil harbours have increased the risk of oil-spills. Between 1992 and 2003, the total hydrocarbon concentrations (THCs) of Baltic subsurface water were determined by fluorescence spectroscopy (UVF). Concentrations ranged from 0.13 to 1.8 μg l^{-1} as Ekofisk crude oil equivalents. To assure comparability of the results, the values were converted to chrysene equivalents (0.010–0.14 μg l^{-1}). Seasonal dependence of the THCs was evident — the lowest concentration was found during summer in the Bothnian Sea and Bothnian Bay and the highest during winter in the northern Baltic Proper. THCs differed geographically following the amounts of oil discharges, with highest values in the Gulf of Finland, the northern Baltic Proper and the eastern Gotland Basin.

Introduction

Petroleum hydrocarbons in the marine environment may originate from a variety of sources: fossil fuels, municipal and industrial wastes, runoff, oil accidents, illegal discharges and shipping. Oil and oily discharges from ships represent a significant threat to marine ecosystems. These discharges may be intentional or accidental. Intentional illegal discharges of oil from ships tend to be limited in size and geographically scattered, but, surprisingly their sum is greater than the amounts in oil spills, and they may create a chronic impact in certain areas. Historically, accidental oil spills have been of crude oil rather than of refined products such as diesel or fuel oil, but in recent years the trend has reversed and spills of heavy fuel oil are more common. Although there have been no large oil spills from tankers during recent years, oil is considered a continuing threat to the Baltic Sea ecosystems. HELCOM (the Helsinki Commission) has estimated that 20 000–70 000 tonnes of oil enter the Baltic Sea every year and 10% of the total amount comes from illegal discharges from flushing of machinery systems or cargo tanks of vessels (HELCOM 2003 and http://www.helcom.fi/manandsea/shipping/oilpollution.html). During 2001, surveillance aircraft detected about 400 illegal oil discharges in the Baltic Sea (HELCOM 2003 and http://www.helcom.fi/manandsea/shipping/aerialsurveillance.html). In most cases the amounts discharged were estimated at less than one cubic metre. Not all spillages are observed, however.

The probability of shipping accidents has increased with the heavier maritime traffic on the Baltic Sea. Oil terminals are now operating in Finland, Russia, Estonia, Latvia and Lithuania and new ones are under construction or in the planning stage. Shipping in the Baltic Sea accounts for about 15% of all maritime traffic around the world. The amount of oil transported in the Baltic Sea (80 million tonnes in 2000) is expected to increase up to more than 130 million tonnes per year by 2015. Increasing oil transports will increase the risk of large oil-spills, involving over 10 000 tonnes of oil, by 35% for the whole of the Baltic Sea and 100% for the Gulf of Finland (Rytkönen *et al*. 2002, HELCOM 2003).

Crude oil and other fossil fuels contain polycyclic aromatic hydrocarbons (PAHs). Crude oil and oily wastes in general undergo a number of physical and chemical alterations (called weathering) in the marine environment. Evaporation, photooxidation, metabolic degradation and other processes interact to reduce PAH concentrations in water (Neff 1979). Ultra-violet fluorescence (UVF) is considered to be a highly sensitive, reasonably selective, simple, rapid and straightforward method to determine oil-based aromatic compounds in seawater (IOC 1984). The method is based on the similarities between the fluorescence excitation and emission spectra of non-polar organic compounds extracted from seawater and those present in most refined and crude oils (Knap *et al*. 1986).

Some analytical data for total petroleum hydrocarbons (THCs) in the Baltic Sea subsurface water are available from the past (Tervo 1978, Tervo 1980, Law and Andrulewicz 1983, Poutanen and Tervo 1987, Poutanen 1989, Poutanen *et al*. 1990, HELCOM 2002). The purpose of the present paper is to evaluate the total hydrocarbon content of Baltic Sea subsurface water between 1992 and 2003, concentrating on waters close to Finland. The UVF screening method with Ekofisk crude oil as standard oil was used as the basis of comparison. The UVF method performance characteristics are presented. The relation between Ekofisk standard oil and chrysene was determined to allow comparability of the results with other data sets.

The fluorescence excitation and emission wavelengths that were used — 310 nm and 360 nm, respectively — measure the complex mixture of compounds containing two or more aromatic rings (IOC 1984). Three- and four-ring aromatics emit most strongly between 340 and 380 nm (Law 1981). Some criticism of the UVF technique for the assessment of oil pollution has been presented and, as a consequence, there is no HELCOM database on petroleum hydrocarbon contaminants measured by UVF (HELCOM 1996). The UVF method is valid for screening purposes, however, and especially for field measurements.

Material and methods

Analytical procedures recommended by the Intergovernmental Oceanographic Commission (IOC 1984) were adopted and slightly modified. Fluorescence excitation and emission wavelengths — 310 nm and 360 nm, respectively — were used in the measurements with a luminescence spectrometer (Perkin Elmer LS 50). Ekofisk crude oil (light ends removed, crude oil provided by Bundesamt für Seeschiffahrt und Hydrographie Labor Sülldorf Chemie des Meeres, Germany) was used as calibration standard. A multipoint linear calibration curve was determined for a concentration range of 0–2.6 μg l^{-1}. Chrysene powder (CRM 269, made by BCR, European Commission) was used in the standard comparison study.

Sampling

Two separate subsurface water samples were collected at a depth of 1 m from the foredeck of a research vessel as the vessel approached the sampling station. Sample bottles were carefully cleaned and purity-checked with hexane before use. The mouth of the 1-litre sample bottle made of glass was covered with aluminium foil and a screw-top cap with a hole in it. The sample bottle was placed in a specially designed bottle holder (Tervo 1978). For sampling, the aluminium foil was broken with a “bullet” (Tervo 1978) and the bottle was filled with water. Selected sampling events in 1992–2003 at each sampling station are presented in Table 1.

Sample handling

Part of the sample water was immediately removed from the bottle to obtain a total volume of one litre. Hexane (10 ml, HPLC grade, Rathburn) was added to the water and the bottle was covered again with aluminium foil and a screw-top cap. Extraction was performed with magnetic stirring for 45 minutes. After extraction, purity-checked water (extracted several times with hexane) was added to the sample bottle until the hexane phase was well separated. Hexane was withdrawn with a pipette to the quartz cuvette for immediate total fluorescence measurement.

Results

Total hydrocarbon concentrations (THCs) as Ekofisk crude oil equivalents

THC levels were determined in several areas of the Baltic Sea in 1992–2003: Bothnian Bay, Bothnian Sea, Åland Sea, Gulf of Finland, northern Baltic proper, eastern Gotland Basin and western Gotland Basin (Fig. 1). In addition, several water samples were collected from the southern Baltic Proper and Kattegat in winter 2002. Duplicate samples were taken at each sampling station. The difference allowed between concentrations of the two samples was 25%, calculated by taking the difference between two concentration values and dividing the result by the concentration of the sample with higher value. If the difference was more than 25%, only the lower value was used in comparison. Otherwise the average of the two results was used.

Two average THC values for the years 1992–2003 were calculated for each of the 12 sampling stations, one for summer (from May to September) and one for winter (from December to March). The average of the summer and winter THC values was then calculated, to give a single value for each sampling station. These values ranged from 0.39 to 0.76 μg l^{-1} in the different sea areas (Fig. 1). The values in the different sea areas varied in summer between 0.13 and 0.77 μg l^{-1} and in winter between 0.26 and 1.8 μg l^{-1} (Figs. 2 and 3). Yearly summer and winter THC values for seven of the sampling stations are presented (Figs. 4–10). A comparison of the seasonal THC levels at three sampling stations, located in the Bothnian Sea, the Gulf of Finland and the eastern Gotland Basin, is presented (Figs. 11 and 12).

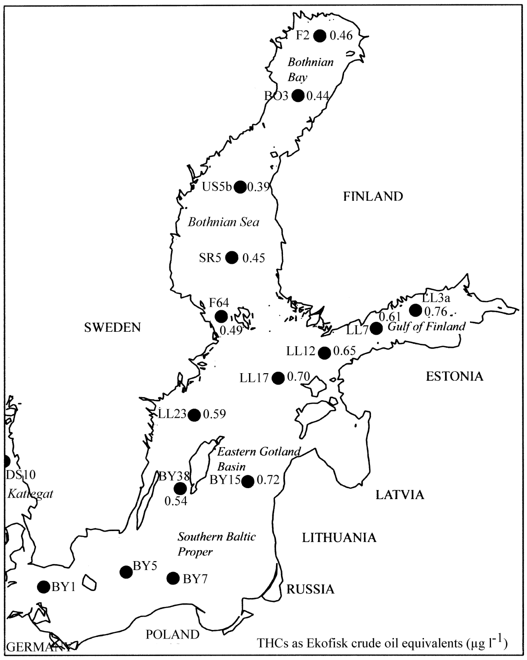

Fig. 1. Map of water sampling sites in the Baltic Sea. Average THCs as Ekofisk crude oil equivalents (µg l^{-1}) in different areas in 1992–2003.

THCs were on a higher level during winter than during summer (Figs. 2–12). Levels were

Table 1. Description of water sampling stations in the Baltic Sea 1992–2003.

Area	Sampling station	Number of THC results (summer/winter)
Bothnian Bay	F2	9/4
Bothnian Bay	BO3	10/5
Bothnian Sea	US5b	11/9
Bothnian Sea	SR5	11/6
Åland Sea	F64	11/9
Gulf of Finland	LL3a	12/7
Gulf of Finland	LL7	12/8
Northern Baltic Proper	LL12	12/7
Northern Baltic Proper	LL17	10/8
Northern Baltic Proper	LL23	7/7
Eastern Gotland Basin	BY15	9/8
Western Gotland Basin	BY38	4/4

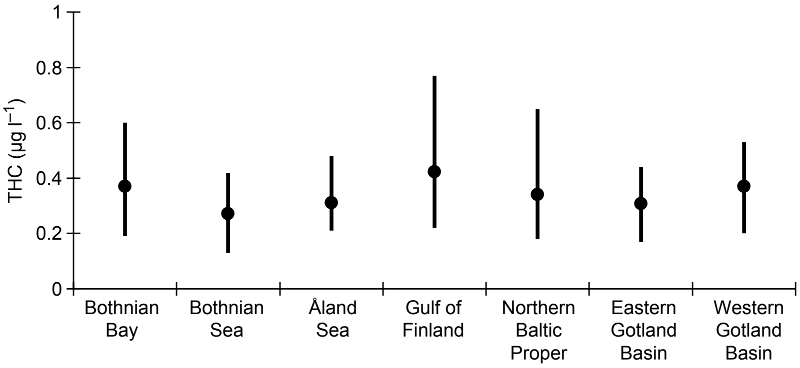

Fig. 2. Total hydrocarbons (µg l–1) in the Baltic Sea water in seven areas during summer in 1992–2003.

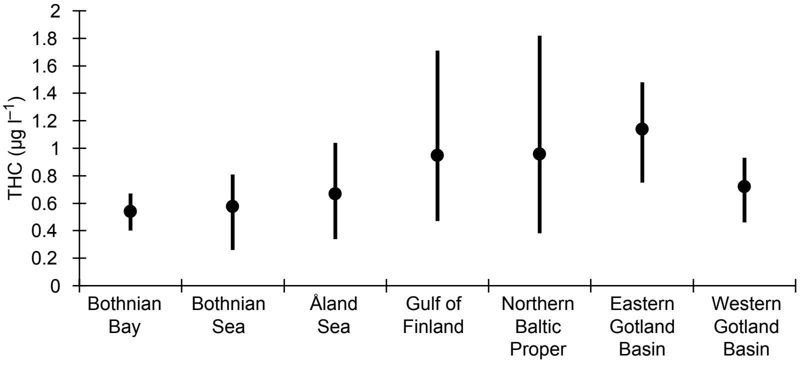

Fig. 3. Total hydrocarbons (µg l–1) in the Baltic Sea water in seven areas during winter in 1992–2003.

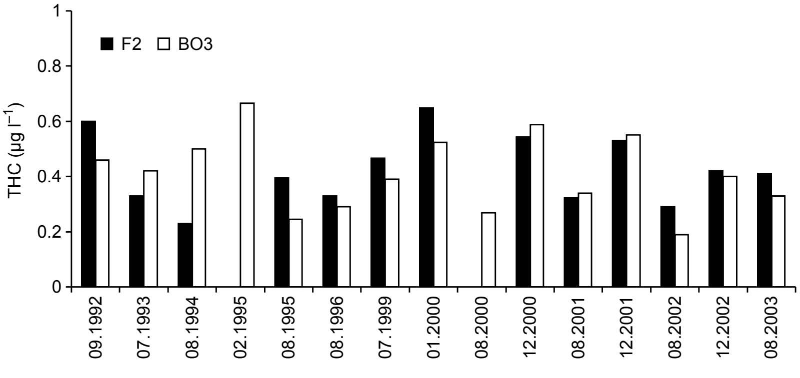

Fig. 4. Total hydrocarbons (µg l–1) in the Bothnian Bay water in 1992–2003. Sampling month and year presented.

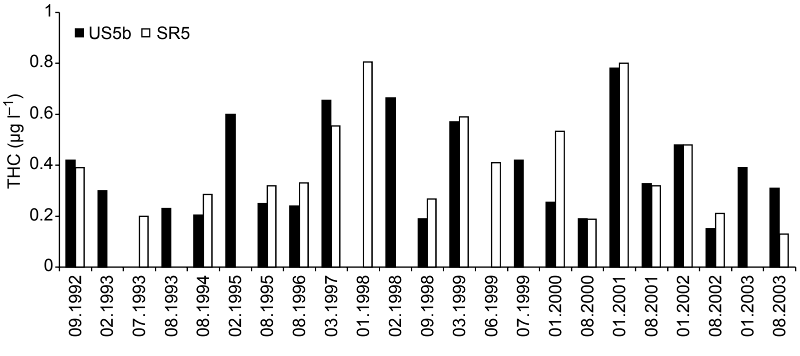

Fig. 5. Total hydrocarbons (µg l–1) in the Bothnian Sea water in 1992–2003. Sampling month and year presented.

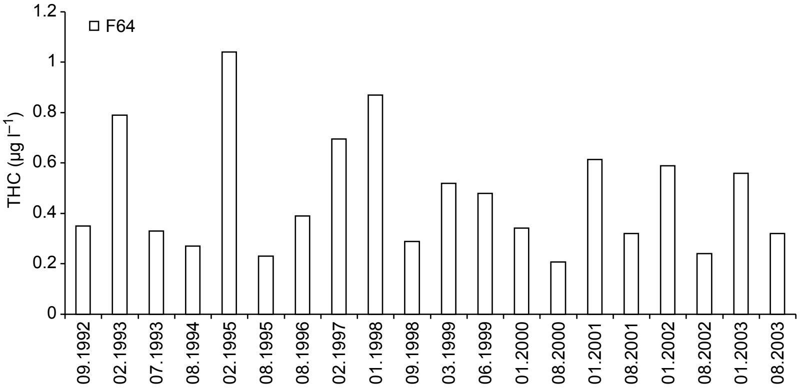

Fig. 6. Total hydrocarbons (µg l⁻¹) in the Åland Sea water in 1992–2003. Sampling month and year presented.

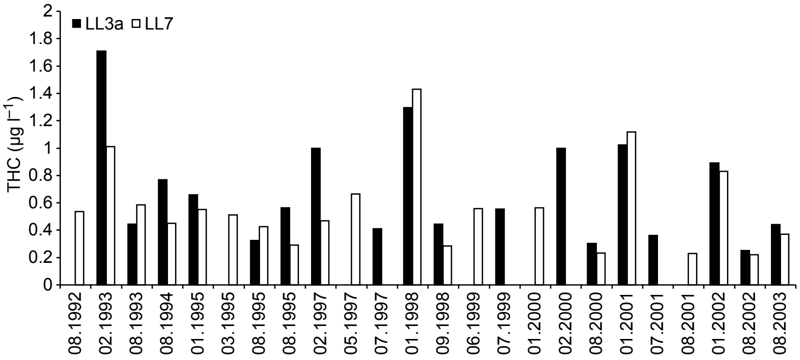

Fig. 7. Total hydrocarbons (µg l⁻¹) in the Gulf of Finland water in 1992–2003. Sampling month and year presented.

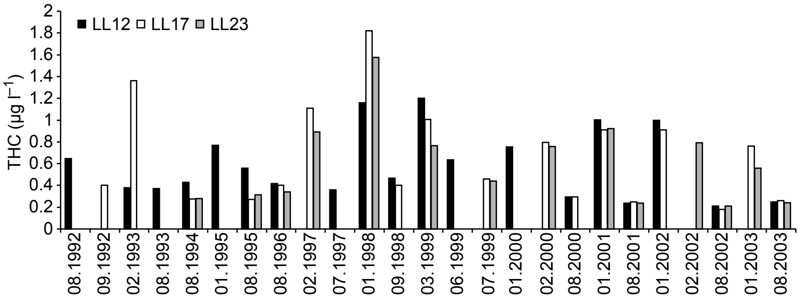

Fig. 8. Total hydrocarbons (µg l⁻¹) in the northern Baltic Proper water in 1992–2003. Sampling month and year presented.

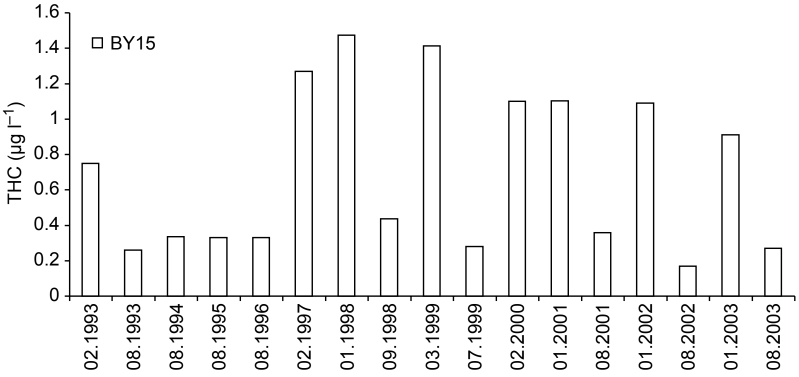

Fig. 9. Total hydrocarbons (µg l⁻¹) in the eastern Gotland Basin water in 1992–2003. Sampling month and year presented.

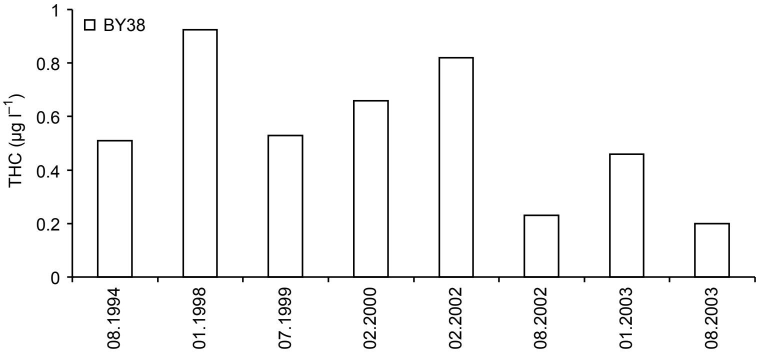

Fig. 10. Total hydrocarbons (µg l^{-1}) in the western Gotland Basin water in 1992–2003. Sampling month and year presented.

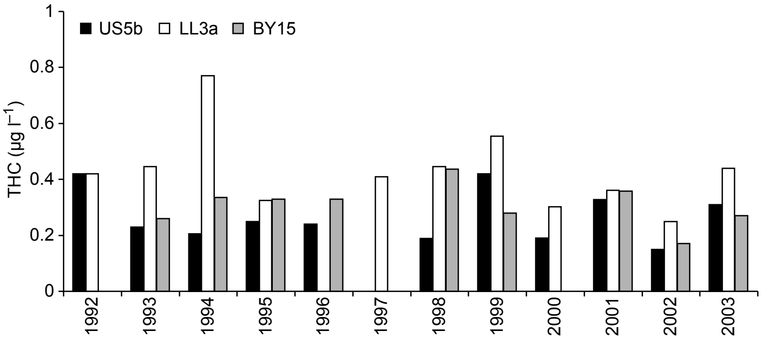

Fig. 11. Comparison of total hydrocarbon levels (µg l^{-1}) in the Bothnian Sea (US5b), the Gulf of Finland (LL3a) and the eastern Gotland Basin (BY15) during summer.

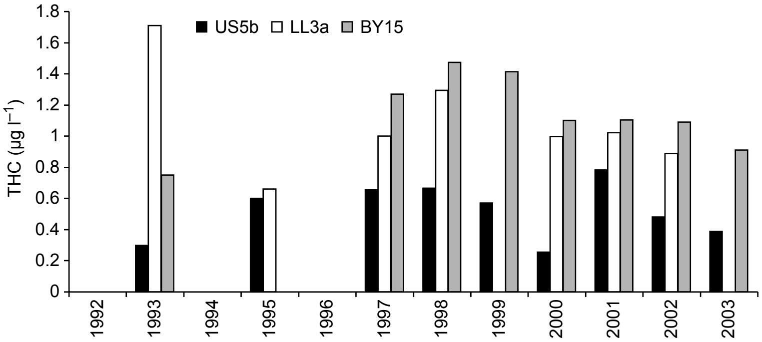

Fig. 12. Comparison of total hydrocarbon levels (µg l^{-1}) in the Bothnian Sea (US5b), the Gulf of Finland (LL3a) and the eastern Gotland Basin (BY15) during winter.

lowest during summer in the Bothnian Bay and the Bothnian Sea, and highest during winter in the eastern Gotland Basin, the northern Baltic Proper and the Gulf of Finland. The seasonal difference in THC levels was largest in the northern Baltic Proper and the eastern Gotland Basin. Average winter values (Fig. 3) for the past 12-year period indicated an increase in concentration from the Bothnian Bay towards the Bothnian Sea and further via the Gulf of Finland to the northern Baltic Proper and the eastern Gotland Basin. A similar THC pattern was observed for winter values in 2002 (January/February) with values increasing from the most northern part of the Baltic Sea (the Bothnian Sea) towards the Baltic Proper and the eastern Gotland Basin (Table 2). During winter in 2002, THC levels were highest in the Gulf of Finland, the northern Baltic Proper, the eastern Gotland Basin and the southern Baltic Proper and lowest in the Bothnian Sea, Åland Sea and Kattegat. Inspection of the seasonal variation in THCs in the years 1992–2003 at three sampling stations — the one with lowest average value of THC (US5b) and the two with highest values (LL3a, BY15) (Fig. 1) — showed the highest summer THC values (Fig. 11) to occur at LL3a, the most eastern station of the Gulf of Finland, and the highest winter values (Fig. 12) at BY15 in the eastern Gotland Basin. THC levels were lowest at US5b in the Bothnian Sea.

Quality assurance

The fluorescence background of hexane (HPLC grade, Rathburn) was checked with a luminescence spectrometer (Perkin Elmer LS 50) using the excitation and emission wavelength pair 310 nm/360 nm. Before use, the sampling bottles and other essential equipment were cleaned with hexane until they were proved to be uncontaminated. Stability of the calibration curve was checked and controlled regularly by analysing a control solution (Ekofisk crude oil in hexane).

Repeatability of the method was evaluated regularly by sampling a large volume of Baltic water and dividing it into subsamples ($n = 5$–10) for extraction. The relative standard deviation was between 5.5% and 14% in the concentration range of 0.16–1.2 μg l^{-1}.

Recovery of the method was determined by spiking water with Ekofisk crude oil. A large-volume water sample was collected and subsampled (1 l, $n = 9$). Three concentration levels (0.3 μg, 0.6 μg and 1.0 μg of Ekofisk crude oil in hexane) and two spiked samples for each concentration level were prepared. Three of these subsamples were used to determine the "background" concentration. The average recovery for the three concentration levels was 104% ($n = 6$) with relative standard deviation of 22%.

Relation of Ekofisk crude oil and chrysene equivalents

Chrysene (a tetracyclic aromatic hydrocarbon) rather than crude oil was used as standard so that total hydrocarbon concentrations could be presented as chrysene fluorescence units and compared with other data.

A stock solution was prepared by dissolving 10 mg chrysene powder in 100 ml hexane (Rathburn, HPLC grade, 100 ml). A series of standard solutions was prepared to determine a calibration curve for chrysene with the same intensity area as for Ekofisk crude oil. The intercomparison ratio R (IOC 1984) was found to be 13 calculated from the chrysene and Ekofisk oil calibration lines. Thus Ekofisk crude oil based THCs could be presented as chrysene-based THCs by dividing the Ekofisk crude oil based results by the factor 13.

Discussion

Comparison of the maximum THC values during the past 12 years (Figs. 2 and 3) shows an increase in concentrations from north to south: the Bothnian Bay towards the Bothnian Sea and further via the Gulf of Finland to the Baltic Proper. The increase is compatible with the greater number of oil spills towards the south (http://www.helcom.fi/manandsea/shipping/aerialsurveillance.html).

The range of summer THC values in 1992–2003 in the eastern Gotland Basin (0.17–0.44 μg l^{-1}) (Fig. 2) was on a lower level than the range in the western and central parts of the Baltic Sea (0.5 to 1.6 μg l^{-1}) (HELCOM 2002) during the summer months of 1997 and 1998. In our studies, no samples for THC analysis were collected from the eastern Gotland Basin during summer 1997 and 1998. Witt and Siegel (2000) found a significant increase of PAH concentrations in southern Baltic surface water in August 1997 (the Oder River estuary) after the

Table 2. Total hydrocarbons in the Baltic Sea in January/February 2002.

Area	Sampling station	THC (µg l^{-1})
Bothnian Sea	US5b	0.48
Bothnian Sea	SR5	0.48
Åland Sea	F64	0.59
Gulf of Finland	LL3a	0.89
Gulf of Finland	LL7	0.83
Northern Baltic Proper	LL12	1.01
Northern Baltic Proper	LL17	0.91
Northern Baltic Proper	LL23	0.79
Eastern Gotland Basin	BY15	1.09
Western Gotland Basin	BY38	0.82
Southern Baltic Proper	BY7	1.06
Southern Baltic Proper	BY5	0.84
Southern Baltic Proper	BY1	0.97
Kattegat	DS10	0.68

highest river flood level in the last 50 years. These increased concentrations were attributed to flooding of municipal and industrial waste disposal areas in the drainage basin of the river (Witt and Siegel 2000). The range of winter values (0.75–1.48 μg l^{-1}) found in the open sea of the eastern Gotland Basin in 1992–2003 is lower than the wintertime concentrations reported by HELCOM (2002), which ranged from 1.1 to 3 μg l^{-1} during the period of 1994–1998. Higher values are indeed reported for coastal regions such as the inner bights and the mouth of the Oder (HELCOM 2002). Our highest value for the eastern Gotland Basin was 1.48 μg l^{-1}, found in January 1998 (*see* Fig. 9). The THC values were also high in other areas in January 1998 (Figs. 5–8 and 10).

The average THCs in 1992–2003 (0.39–0.76 μg l^{-1}) were close to average levels, 0.5–0.7 μg l^{-1} (Ekofisk crude oil equivalents), in 1980–1986 reported by Poutanen and Tervo (1987). In our data for 1992–2003, the range of the maximum values in the Bothnian Bay, the Bothnian Sea, the Gulf of Finland and the northern Baltic Proper (0.7–1.8 μg l^{-1}) was somewhat lower than the maximum values (1.4–2.2 μg l^{-1}) for the same areas in the 1980s (Poutanen and Tervo 1987). The THC levels were higher in the Gulf of Finland in 1987 because of an oil-spill in February in which 570 tonnes of crude oil was released. In May, three months after the spill, just after the ice had melted and when widespread oil films were observed in the Gulf, a maximum value of 14 μg l^{-1} (as Ekofisk crude oil equivalent) was determined in the open sea area of the Gulf of Finland (Poutanen *et al.* 1990). This value is almost eight times the highest level found during 1992–2003. By August 1987 the concentrations had returned to background levels (Poutanen 1989, Poutanen *et al.* 1990).

Unpolluted river water and seawater contain less than 0.1 μg l^{-1} total PAH, whereas total PAH concentrations in river water flowing through heavily industrialised regions range from 1 to 5 μg l^{-1} (Kennish 1997). After the Sea Empress oil spill on the south coast of Wales in 1996, background concentrations of total petroleum hydrocarbons for the region were estimated to be 0.2–0.9 μg l^{-1} in seawater (OSPAR Commission 2000). THC levels below 1 μg l^{-1} are considered typical of seawater without significant petroleum pollution (Law 1981, Bícego *et al.* 1996, Zanardi *et al.* 1999). In the present study, covering the years 1992–2003, all the THC levels during summer were above 0.1 μg l^{-1} but below 1 μg l^{-1}, whereas during winter the level of 1 μg l^{-1} was exceeded in many areas of the Baltic Sea (Åland Sea, Gulf of Finland, northern Baltic Proper, eastern Gotland Basin, southern Baltic Proper). The maximum values were below 1.8 μg l^{-1}, however, which is well within the range of 1 to 5 μg l^{-1}, which is mentioned as the range for total PAH concentrations in river water flowing through heavily industrialised areas. During 1992–2003, THCs were within the range of 0.13–1.8 μg l^{-1} as Ekofisk crude oil equivalents, which is slightly lower than the level 1.1–3.5 μg l^{-1} (Ekofisk crude oil equivalents) reported by Law (1981) in UK marine waters. In June 1993, a range of 0.2–0.5 μg l^{-1} (Ekofisk crude oil equivalents) was found (Kirby *et al.* 1998) in offshore sites in UK marine waters. The level is close to the range of summer values (0.13–0.77 μg l^{-1}) found in our studies for 1992–2003.

The whole THC range found in Baltic water in 1992–2003 was determined to be 0.010–0.14 μg l^{-1} as chrysene equivalents. Using as the basis of comparison the average yearly values found in the Baltic Sea, with the range 0.39–0.76 μg l^{-1} as Ekofisk equivalents (Fig. 1) and converting to chrysene equivalents, the values would be 0.030–0.058 μg l^{-1}. That is a higher level than for the ocean water samples collected off Bermuda, 0.011 μg l^{-1} as chrysene equivalents (n = 44, RSD 65%) (Knap *et al.* 1986). However, a higher level, 0.057 μg l^{-1} as chrysene equivalents (n = 30, RSD 60%), is reported for the inshore waters off Bermuda (Knap *et al.* 1986), and this value is close to the values found here in the open sea Baltic water. For the eastern Mediterranean Sea, Yilmaz *et al.* (1998) report a range of 0.01–4.14 μg l^{-1} as chrysene equivalents — with mean values between 0.20 and 0.35 μg l^{-1} — in surface water (determined in July, September and October 1995). This range is considerably higher than our summer range of 0.010–0.059 μg l^{-1} as chrysene equivalents in 1992–2003. Shriadah (1999) reports petroleum hydrocarbon levels in surface water ranging from 0.75 μg l^{-1} up to 12.95 μg l^{-1} (as chrysene equivalents). These much higher concentrations were found in 1993–1994 in the offshore stations off the Arabian Gulf coast of the United Arab Emirates close to the busiest tanker routes in the world.

In our study, there was a clear seasonal difference in the THC levels of Baltic Sea surface water: the THC levels were higher during winter than summer. There are several processes — volatilisation, photooxidation, chemical oxidation and metabolism — that remove petroleum hydrocarbons from seawater. The degradation rate depends on the environmental conditions. During the cold season, degradation is minimised because of the reduced temperature and light intensity, and THC levels are higher. Also cold conditions slow down the microbial decomposition of petroleum hydrocarbons. In warmer conditions with more sunlight, petroleum hydrocarbons volatilise more readily and undergo photooxidation and THC levels drop. THC levels of seawater are also lower after spring/summer phytoplankton blooms, when detritus settles down to the sea bottom, withdrawing petroleum hydrocarbons (PAHs) that have been absorbed by phytoplankton (Kowalewska and Konat 1997, Kowalewska *et al*. 2003). It is reported by EEA (2003) that phytoplankton biomass expressed as chlorophyll *a* is increasing at a few stations in Finnish, Lithuanian and Swedish coastal waters. Although the process of sorption of PAHs is complex and is dependent on many parameters, phytoplankton, especially during blooms, appears to clean the water in the southern Baltic ecosystem and to transport PAHs to the sediment (Kowalewska 1999). The effects of oil hydrocarbons on Baltic Sea ecosystems are diverse with possible acute toxic effects and long-term effects via accumulation of hydrocarbons on biota and sediments.

Conclusions

The geographical and seasonal THC trends in Baltic subsurface water during 1992–2003 parallelled the location of oil spillages and indicated an increasing level of pollution from north to south. Overall, no marked changes in levels were evident during the last twelve years. The relationship determined between Ekofisk crude oil and chrysene equivalents made the results widely comparable. The UVF technique that was employed is a suitable screening technique for the evaluation of the THC levels in Baltic Sea water. Its value is emphasised during unusual events when information about normal THC levels is of importance. The THC results presented describe well the overall development in the long-term and provide an excellent baseline against which to compare THCs after possible oil accidents in the future.

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