WEATHERING OF TANKER PRESTIGE AND ERIKA FUEL OILS N°6 IN MARINE ENVIRONMENT[1]

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ABSTRACT

The shipwreckage of the oil tanker Prestige and Erika occurred respectively on November the 19th 2002 off the coast of Galice in Spain and on December the 12th 1999 in the Golf of Gascogne. Both led to large oil spills that contaminated hundreds of kilometres coast: more than 10 000 tons of fuel oil n°6 were released in the Atlantic Ocean in the south of Finistère as a consequence of the Erika and probably four times more from the Prestige east of Galicia.

As for the product transported by the tanker Erika, the fuel n°6 of the Prestige was little sensitive to the action of dispersing agents. The water-in-oil emulsion formed during its contact with water led to large sheets that floated and drifted under the sea surface in the direction of the coast. Samples reaching the coast after different residence times in the ocean were analysed and the results were compared to those of the reference product charged. The results showed that the fuel composition were similar in both products.

For both fuels, laboratory experiments have been performed in closed systems at 20°C in order to evaluate the maximum transfer of hydrocarbons to seawater with slow agitation of the water, ensuring a constant homogenisation of the aqueous phase.

For both fuels, the hydrocarbon solubilisation at equilibrium after 2 days were very similar. The concentration of BTEX solubilised in seawater was between 210 and 260 µg/L for Erika fuel and 185 µg/L for the Prestige.

The concentration of total hydrocarbons was about 560 to 750 µg/L for Erika fuel and 570 µg/L for the Prestige. At the same time, the concentration of solubilised polyaromatic hydrocarbons (according the list of the US-EPA) was from 140 to 190 µg/L for Erika fuel and 133 µg/L for the Prestige.

Biodegradability tests of these fuel oil n°6 have been performed using a hydrocarbon degrading microflora whose intrinsic capacity to degrade hydrocarbons has been checked in previous works. In both cases, it appeared that 9% of the maltenes fraction could be degraded under optimised laboratory conditions, linear alkanes below nC_{40} were nearly totally biodegraded, 98% for Erika and 92 % for Prestige.

Concerning the biodegradation of the saturated and aromatic fractions, the biodegradation behaviour is comparable. Even though some differences exists that could be attributed to the respective composition of each petroleum cut. The biodegradation ratio of the total fraction of saturated hydrocarbons was about 21% for Erika and 61% for the Prestige. The polyaromatic hydrocarbons (PAH) with less than four rings were completely biodegraded for Erika while 85% were biodegraded for the Prestige. With 70% and 66 % respectively, the fluoranthene biodegradation are similar. The pyrene concentration, degraded at 25% for Erika, remain constant for the Prestige after 80 days. For higher molecular weight PAH, no significant disappearance could be noticed.

INTRODUCTION

The weathering of the petroleum hydrocarbons spilled into the sea is due to several physical, chemical and biological processes. The modification of their composition by evaporation, dissolution, dispersion, and biodegradation may induce negative impacts on the environment.

The aim of this work was to present data on the off shore evolution and the potential environmental impact of the fuel oil n°6 charged in the Erika and the Prestige tanker. For that purpose samples taken after different residence times in the ocean have been analysed and the results were compared to those of the reference product. Complementary laboratory experiments were realised in order to determine the aqueous transfer of the hydrocarbons contained in the fuel and their biodegradability.

MATERIAL AND METHODS

SARA fractionation method covers the separation and the quantitative measure of the four generic fractions inherently present in all petroleum crude oil. The 4 fractions are saturated hydrocarbons, aromatic hydrocarbons, resins and asphaltènes. The asphaltenes are quantified by gravimetry after precipitation of the non soluble fraction in n-hexane at 20°C. The global soluble fraction in n-hexane or maltenes fraction is quantified either by gravimetry or by GC/FID analysis. The fractionation is operated by liquid chromatography in a glass column filled with activated silica (70-230 mesh). Solvents of increasing polarity are used to elute saturates, aromatic and resin fractions quantified by gravimetry or by GC/FID analysis.

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[1] Fuel oil n°6 is equivalent to fuel oil n°2 according to European specifications.

Gas chromatographic analysis of Erika's oil were performed on a commercial available 3500 Series Varian Gas Chromatograph equipped with a flame ionisation detector (FID). Separation of C10/C40 hydrocarbons is achieved using a fused DB5 silica capillary column of 60m length, 0.25 mm i.d, 0,1 µm film thickness. The programmed oven temperature applied to the sample started at 35°C till 310°C at 3°C/min.

High temperature gas chromatographic analysis of Prestige's oil were performed on a commercial available Trace GC 2000 Series Thermofinnigan Gas Chromatograph equipped with a flame ionisation detector (FID). Separation of C10/C70 hydrocarbons is achieved using a fused MXT-1 Crossbond silica capillary column of 6 m length, 0.53 mm i.d, 0,15 µm film thickness. The programmed oven temperature applied to the sample started at 35°C till 350°C at 10°C/min.

The Pollut-Eval ä is a fully automatic system of pyrolysis and oxidation for the characterisation of hydrocarbons in contaminated soil. It allows a fast analysis of soil samples without any solvent extraction and can also be involved in the analysis of pure hydrocarbons. The treatment applied to the sample consist of two successive step. In a first time the sample is introduced in the pyrolysis oven flushed by nitrogen, the hydrocarbons are thermovaporised and then cracked by heating from 80°C to 650°C. The heating rates (25°C/min) are adjusted so as to release the various petroleum cuts which are measured by a flame ionisation detector (FID). Then, in the oxidation oven, the samples are oxidised under air flow from 350 to 850°C. The oxidation of the residue allows to calculate the total organic carbon of the sample. During this two steps an infra-red detector measures continuously the CO and CO_2 released. This approach gives the complete carbon mass balance of the sample.

Diphasic water/oil contact are realised at 20°C in closed vessels. 10 g of oil and 2 L of reconstituted seawater are stirred with a slow agitation to ensure a constant homogenisation of the aqueous phase without any formation of oil droplets. During 7 days, various water sampling are analysed to determine the kinetics of the hydrocarbon transfer into the aqueous phase.

Total dissolved hydrocarbons and BTEX analysis are performed on a 3400CX Series Varian Gas Chromatograph equipped with a flame ionisation detector (FID) and a Purge & Trap injector (Tenax trap). Separation of hydrocarbons is achieved using a fused Petrocol DH silica capillary column of 100 m length, 0.25 mm i.d, 0,5 µm film thickness. The programmed oven temperature applied to the sample started at -40°C till 210°C at 2°C/min.

Polyaromatic Hydrocarbon analysis of the US-EPA priority pollutant list are performed with a Waters Millenium HPLC equipped with a LC PAH column of 25 cm length, 4.6 mm diameter, 5µm. Separation of the 16 PAH is performed under water/acetonitrile gradient at 1mL/min. The quantification is performed with aWaters 996 photodiode array detector and a Waters 470 scanning fluorescence detector.

Biodegradability test are performed during 14 weeks in a Sapromat respirometer after inoculation of a mixture of bacterial population which intrinsic ability to degrade the hydrocarbons has been previously described (Haeseler et al. 1999 and Blanchet D.). The biodegradation tests have been realised at 20°C in synthetic seawater described below.

Component	Concentration (g/L)
NaCl :	23,9
$MgCl_2$; $6\,H_2O$:	10,8
$CaCl_2$:	0,10
KCl :	0,74
Na_2SO_4 :	4,00
$NaHCO_3$:	0,196
K_2HPO_4 :	0,07
NH_4Cl :	0,90
Mineral trace compound solution:	10 mL/L
pH 8,2 with KOH 5N	

Mineral trace compound solution pH2 with HCl	
H_3BO_3	23 mg/L
NaF	2 mg/L
SiO_2, Na_2O, $5H_2O$	22 mg/L
$FeSO_4$;$7\,H_2O$	2 mg/L

The stirring of the media was realised by the respirometeric apparatus Sapromat that supplied permanently the reactor with oxygen avoiding any loss by volatilisation. The strong and constant agitation insured optimal accessibility for the microflora to the hydrocarbons.

RESULTS

The weathering of the fuel oil n°6 of the Erika and the Prestige tanker has been evaluated by two complementary approaches: (1) laboratory analyses performed on samples taken in marine environments in order to assess the evolution of the hydrocarbon composition in the fuel, (2) laboratory experiments realised to determine both the maximal transfer from hydrocarbons in the aqueous phase and the maximal biodegradability of the fuel.

Laboratory analyses on environmental samples

A fractionation into saturated hydrocarbons, aromatic hydrocarbons, resins and asphaltenes has been done on a sample that have been taken off shore by the ship Ailette some days after the shipwreckage.

The results of these analyses are presented in table 1; they were compared to those of the reference product transported in the respective tanker. It can be noted that the proportion of each hydrocarbon family of the 2 original fuel oil n°6 are very close: aromatic hydrocarbons representing 54% to 55% of the fuel with 25% to 23% of saturated hydrocarbons.

Despite their contact with water, the results of the weathered samples taken after the shipwreckage (several days drift in the sea) appeared to be very close to the results obtained with the fuel transported in the tankers. Because of the winter conditions no significant photo-oxidation effect could be noticed in the samples taken offshore (Haeseler et al. 2002).

table 1: Fractionation of different Erika fuel samples into saturated hydrocarbons, aromatic hydrocarbons, resins and asphaltenes.

Proportion (%)	Erika		Prestige	
	Cargo	Offshore sampling	Cargo	Offshore sampling
Saturated HC	25	24	23	21
Aromatic HC	55	54	54	54
Resins	13	16	12.5	11.3
Asphaltènes	7	6	10.3	10.3

The GC/FID simulated distillation of the two original fuel transported by the Erika and the Prestige tanker led to the respective analytical profile presented by Fafet et al 2004.

The global hydrocarbon characterisation realised by the Pollut-Eval method showed that the pyrolysis profile are similar for both the non weathered fuels (figure 1and figure 2). The sample that stayed for a long time in the water (Pornic), showed in the first part of the pyrogram, a lost the lightest fraction. This loss can be attributed to volatilisation, solubilisation and/or biodegradation. The same loss could not be noticed between the Prestige oil and the sample arrived in Mimizan, both of them presented very similar pyrograms (figure 2).
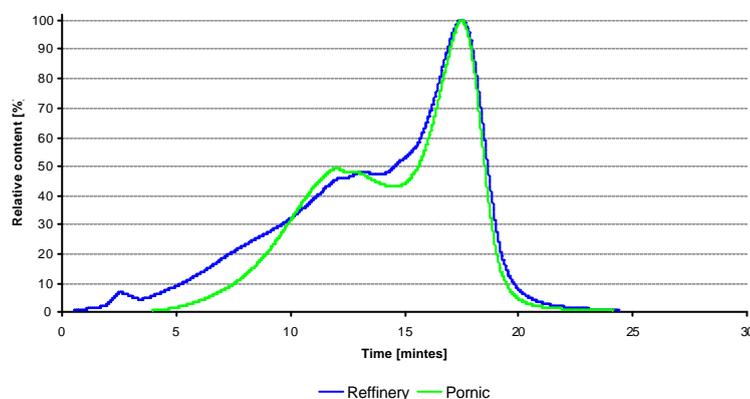
figure 1: Pollut Eval Pyrograms of the Erica cargo and the fuel sampled in Pornic the 26th of August 2000.

figure 2: Pollut Eval Pyrograms of the fuel n°6 of the Prestige cargo and a sample originating from Mimizan.
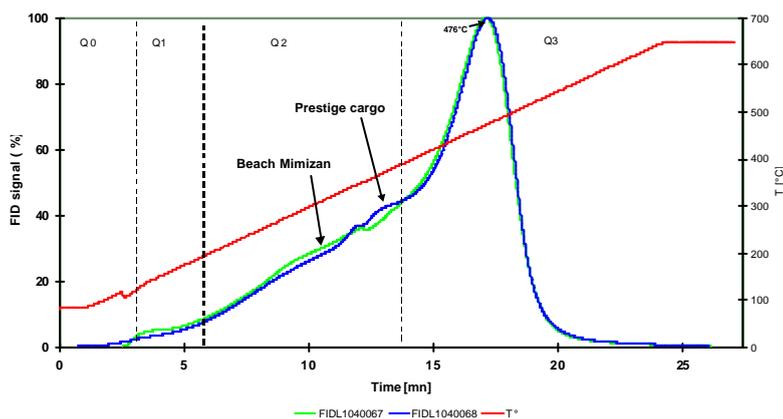
Laboratory experiments

Aqueous transfer experiments with fuel oil n° 6 originating from Erika and Prestige have been performed in closed systems at 20°C in order to evaluate the maximum transfer of hydrocarbons to seawater under slow agitation by simulating a diphasic system and ensuring a constant homogenisation of the aqueous phase.

Two litres of seawater were set in contact with 10 g of fuel oil floating on the surface. The results of these experiments are presented in table 2 and table 3. The following parameters have been analysed at equilibrium after 2 days:

- Volatile hydrocarbons,
- Polyaromatic hydrocarbons,
- Dissolved organic carbon.

The concentrations for BTEX, volatile dissolved hydrocarbons and PAH observed for the Erika fuel are about 25% higher than those observed for the Prestige fuel at equilibrium (table 3).

The detailed concentrations of the 16 PAH from the priority pollutant list of the US-EPA and the BTEX are presented in the table 2 and led to the same conclusion. Except for acénaphtène, the concentrations of the PAH of the US-EPA list are higher for Erika's fuel. This is probably due to a higher molar fraction of all these components in the original Erika oil. Nevertheless, the order of magnitude of the dissolved hydrocarbon concentration observed for both water/oil contacts are similar and the potential impact on the marine environment and their bioavailability could be considered equivalent.

table 2: Detailed concentrations of PAH components dissolved at equilibrium

Components	PAH concentrations[µg/L]	
	Prestige	Erika
Naphtalène	78	166
Acénaphthylène	1,4	0,8
Acénaphthène	48	3,1
Fluorène	1,8	1,7
Phénanthrène	2,6	9,8
Anthracène	0,2	< 0.01
Fluoranthène	0,7	0,6
Pyrène	< 0.2	0,6
Benz(a)anthracène	< 0.02	< 0.02
Chrysène	< 0.03	0,07
Benzo(b)fluoranthène	< 0.01	< 0.01
Benzo(k)fluoranthène	< 0.23	< 0.23
Benzo(a)pyrène	< 0.14	< 0.14
Dibenz(a,h)anthracène	< 0.02	< 0.02
Benzo(g,h,i)perylène	< 0.01	< 0.01
Indeno(c,d)pyrène	< 0.01	< 0.01
Sum of the 16 PAH (EPA list)	132	183
Sum of the 16 PAH without Naphtalène(US-EPA list)	54	17

The dissolution kinetic, realised for the Erika oil showed that the equilibrium is reached in the water after 2 days contact. At this time the concentration in the aqueous phase is about 900 µg/L for the volatile dissolved hydrocarbons and 250 µg/L for the BTEX (Benzene, Toluene, Ethylbenzene and Xylene).

The same type of experiment has been realised with an Erika oil sample, which has been sampled ashore. The results appeared to be different (Haeseler et al 2002). After reaching the equilibrium the concentration in the aqueous phase was about 156 µg/L for the total hydrocarbons and 32 µg/L for the BTEX. This strong reduction of hydrocarbons transferable to the aqueous phase can be explained by a lower concentration of soluble hydrocarbons in the oil (due to volatilisation and dissolution during the stay in the ocean), but also by the formation of an emulsion which strongly reduces the transfer surfaces.

The order of magnitude of hydrocarbon concentration in the aqueous phase after the same type of diphasic contact with other petroleum product is significantly higher:

- for gasoline, the concentration of total hydrocarbons is about 210 000 µg/L and of BTEX about 160 000 µg/L,
- for kerosene, the concentration of total hydrocarbons is about 15 000 µg/L and of BTEX about 9 000 µg/L,
- for diesel oil, the concentration of total hydrocarbons is about 2 800 µg/L and of BTEX about 1 000 µg/L.

The concentrations measured in this type of diphasic assay must be considered as maximal values that can be reached. In the ocean, the salinity, the much lower temperature and the effect of dilution in this open media contribute to reduce significantly the concentrations of dissolved hydrocarbons originating from the fuel oil n°6. These results cannot be used to infer potential impacts on the environment or the human food chain.

Concerning the oil from Erika and Prestige, it appeared that naphthalene, toluene, meta and ortho xylene were the most concentrated hydrocarbons in the water. Some hydrocarbons like four and five ring PAH were present at very low concentrations in the aqueous phase. According to Raoult's law, this is due to both the low solubility of theses compounds in water and their low concentration in the oil phase (Hemptine de J.C. et al. 1998).

The PAH concentration in the aqueous phase after a diphasic contact with the water in oil emulsion sampled on the beach at the end of December 1999 were much lower than after a contact with the cargo oil. This also emphasised the strongly reduced aqueous transfer due to both fewer surfaces available for contact with water and a rigidified oil phase consecutive to the loss of the light hydrocarbon fraction in the emulsified product.

table 3: Detailed concentrations of volatile dissolved hydrocarbon at equilibrium

Components	Concentration (µg/L)	
	Prestige [1]	Erika [2]
Benzène	38	35
Toluène	62	96
Ethylbenzene	15	19
Meta-xylene	28	58
Para-xylene	11	19
Ortho-xylene	30	38
Isopropylbenzene	0.18	0.6
n-propylbenzene	3.5	2.9
1-methyl-3-ethylbenzene	9.0	18
1-methyl-4-ethylbenzene	4.0	6.2
1,3,5-trimethylbenzene	4.5	9.0
1-methyl-2-ethylbenzene	7.4	9.0
1,2,4-trimethylbenzene	22	49
1,2,3-trimethylbenzene	11	18
1-methyl-4-isopropylbenzene	0.88	1.10
1-methyl-3-isopropylbenzene	0.82	0.33
Indane	6.4	31
1-methyl-2-isopropylbenzene	0.21	0.06
1,3-diethylbenzene	n.d.	2.3
1-methyl-3-n-propylbenzene	1.85	3.5
1-methyl-4-n-propylbenzene	1.28	5.1
1,3-dimethyl-5-ethylbenzene	2.8	0.73
1,4-diethylbenzene	0.85	1.11
1-methyl-2-n-propylbenzene	1.59	1.7
1,4-dimethyl-2-ethylbenzene	3.1	5.4
1,2-dimethyl-3-ethylbenzene	2.4	4.8
1-methylindane	0.07	8.5
1,2-dimethyl-4-ethylbenzene	4.9	0.40
1,3-dimethyl-2-ethylbenzene	n.d.	3.9
1,2,3,5-tetramethylbenzene	2.5	6.7
1,2,4,5-tetramethylbenzene	5.0	10.0
5-methylindane	2.9	21
4-methylindane	5.1	7.0
1,2,3,4-tetramethylbenzene	7.5	30
Tetraline	0.88	6.9
Naphtalene	67	136
2-methylnaphtalene	0.77	0.29
1-methylnaphtalene	14	0.13
Volatile dissolved hydrocarbon	573	748
BTEX	185	256

(1): SPMEGC/FID/HydroCARB analyses

(2): Purge & Trap/GC/FID HydroSTAR analyses.
nd : non detected (concentrations lower than 0,02 µg/L)

Biodegradability tests of the fuel oil n°6 transported by the Erika and the Prestige tanker have been performed in a laboratory respirometer after inoculation of a microflora able to biodegrade the constitutive hydrocarbons.

The time course of the oxygen consumption given by the Sapromat during the biodegradation of the fuel oil is shown in figure 3 for the Erika cargo and in figure 4 for the Prestige Cargo. In both cases, these data indicated that the biodegradation started very rapidly after the beginning. After a logarithmic phase during the first week the oxygen consumption became almost linear and was still going on after 110 days for Erika's oil and 80 days for Prestige oil respectively.
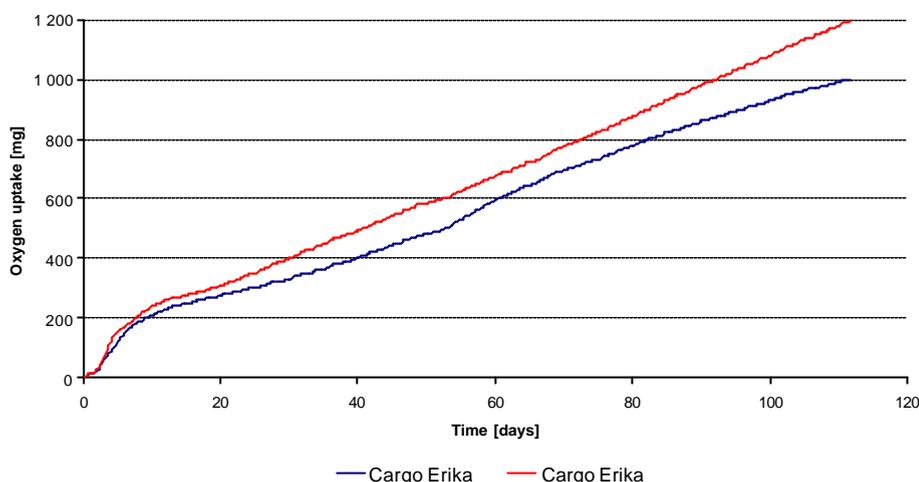
figure 3: Time course of the oxygen consumption given by the Sapromat during the biodegradation of the fuel oil originating from the Erika Tanker.
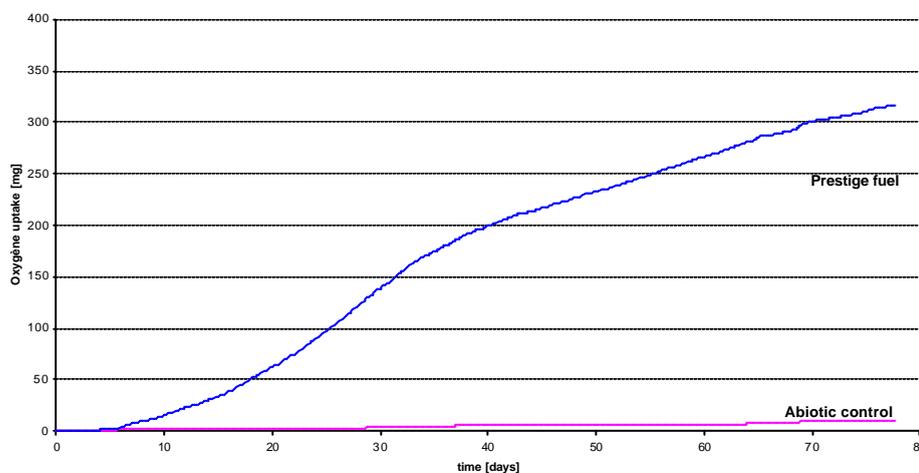
figure 4: Time course of the oxygen consumption given by the Sapromat during the biodegradation of the fuel oil originating from the Prestige Tanker.

The curves presented for the Erika fuel in figure 3 showed an excellent reproducibility of the oxygen consumption in two tests. The same reproducibility was found for the analytical results of residual hydrocarbons after biodegradation.

The analytical results of the biodegradation tests with the Erika fuel oil showed that:
- after 110 days about 9% of the maltenes fraction was biodegraded.
- the polyaromatic hydrocarbons with less than four rings were completely biodegraded while fluoranthene and pyrene, both PAH with four rings, were respectively biodegraded up to 70 and 25 %. For the other PAH from the US-EPA list of priority pollutants no significant biodegradation could be noticed,
- the linear alkanes (ranging from C_{11} to C_{40}) representing 25 g/kg of the initial fuel have been nearly completely biodegraded (up to 98%). For the whole fraction of saturated hydrocarbons the biodegradation level reached about 21%,
- the concentration of resins was noticed to increase during the biodegradation of the fuel oil. This is probably due to the production of polar metabolites, which may have been generated during the incomplete biodegradation of

hydrocarbons and can be quantified as resins due to the analytical procedure used (extraction with methylene chloride).

The analytical results of the biodegradation tests with the Prestige fuel oil showed that:

- a biodegradation level of 9% was observed on the maltenes fraction after 80 days.
- the polyaromatic hydrocarbons (PAH) with less than four rings were not completely biodegraded in 80 days. The biodegradation levels are between 50% for acenaphtene and 85% for naphthalene and phenanthrene. Pyrene was completely recalcitrant to biodegradation..
- the linear alkanes (ranging from C_{11} to C_{40}) have been nearly completely biodegraded (up to 92%). For the whole fraction of saturated hydrocarbons (up to C_{70}) the biodegradation level reached 61%. For the determination of this balance, a C10/C70 high temperature GC/FID has been used in order to evaluate the biodegradation level even for high molecular weight saturated hydrocarbon. This analytical tool was not available for the biodegradation tests of the Erika oil and the different tools used could explain the difference of biodegradation results observed between Erika and Prestige oil for the saturated hydrocarbon.
- the concentration of resins was also noticed to increase during the biodegradation of the fuel oil.

table 4: Biodegradation test on Erika fuel oil: Concentration of C_{34}^- hydrocarbons after different biodegradation times (analysed by gas chromatography with a high-resolution capillary column).

time [d]	Concentration [g/kg]				
	0	6	10	30	80
C_{34}^- n-alcanes	23	2.1	0.5	0.3	0.5
C_{34}^- iso and cyclo-alcanes	72	74	58	47	41
16 EPA PAH	2.9	1.4	1.5	0.9	0.9
C_{34}^- aromatic hydrocarbons	136	149	117	99	100
C_{34}^- hydrocarbons	231	225	175	146	142

These results indicate that fuel oil n°6 (here Erika fuel oil) are mainly composed by heavy compounds which are recalcitrant to biodegradation.

The biodegradation tests with the Erika fuel oil presented in table 4 show 9% of the C_{35}^- hydrocarbons of this oil could be degraded under optimised laboratory conditions (same value as for the Prestige fuel oil). Linear alkanes (nC_{11} to nC_{40}) have been nearly totally biodegraded in both cases. The biodegradation behaviour of the saturated hydrocarbons is different for the 2 fuels. In the case of the Prestige, the biodegradation of the total fraction of saturated hydrocarbons is higher (61%) than for the Erika's fuel (about 21%). The aromatic hydrocarbons were degraded at 18% for Erika's fuel and 6.5% for Prestige's fuel. Nevertheless, concerning the 16 PAH of the US-EPA list, the biodegradation levels observed in both cases are similar (65 and 76% respectively).

These results showed that even for the same global biodegradation level which remain very low for these heavy products, the different composition of the saturated and aromatic fraction led to different biodegradation behaviour.

The high temperature chromatograms presented in the figure 5 and the figure 6 give a global overview of the native oils and of the oils after biodegradation. It appeared very clearly that a limited part of the $C_{10}-C_{45}$ fraction was affected by the biodegradation processes and that the heavy hydrocarbon fraction (a vacuum distillation residue) was not biodegraded.
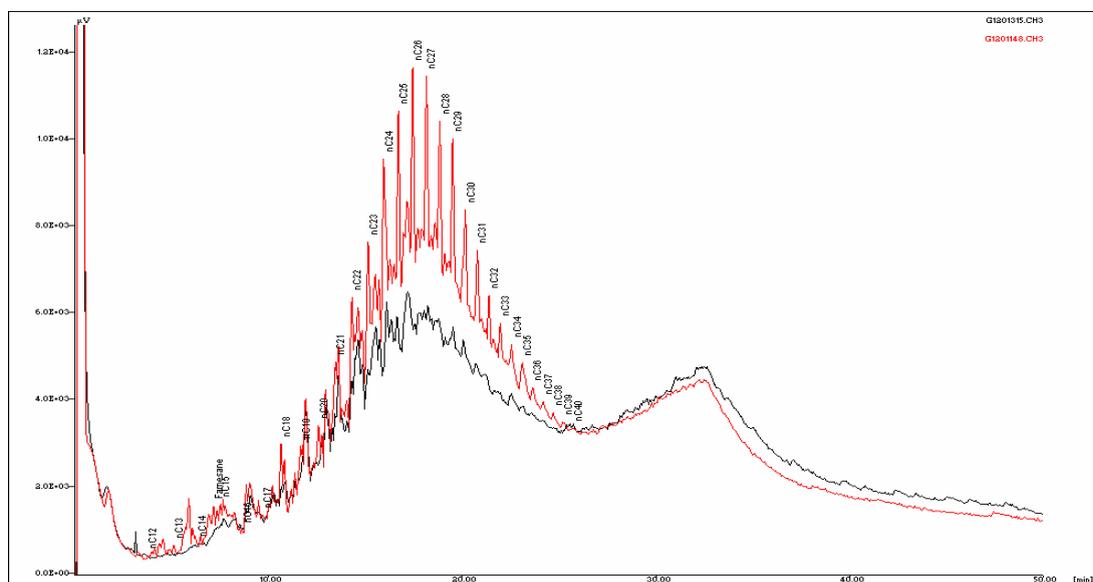
figure 5: High temperature chromatograms of the Erika native oil and the same oil after biodegradation.
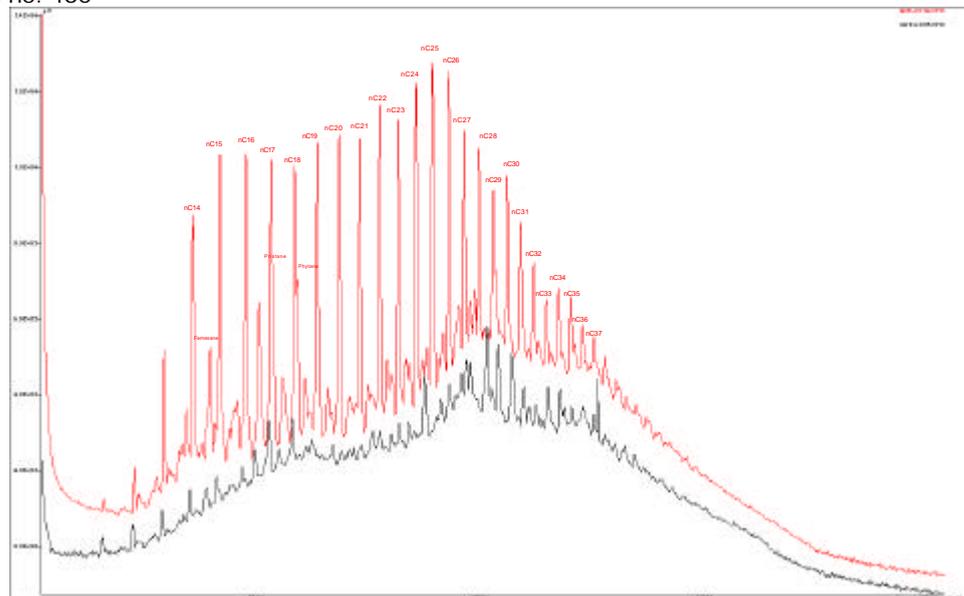
figure 6: High temperature chromatograms of the saturated fraction of the Prestige native oil and after 75 days biodegradation.

CONCLUSION

Among the physico-chemical criteria susceptible to give first indications about the biodegradability of such products, several of them suggested a limited potential for microbiological degradation and transfer to the water phase. From this point of view, the results observed on the fuel oil n°6 transported by the Erika and the Prestige tanker are coherent.

It appeared that the natural attenuation of these oil was not very important in terms of persistence in the environment, but the weathering of these fuel under environmental conditions (emulsification, loss of the volatile fraction, solidifying…) lead to a significant decrease of the impact on the water quality in particular concerning the most hydrosoluble compounds (aromatic hydrocarbons).

It turned out that in both cases, 9% of the initial products could be biodegraded in optimised laboratory conditions. The case of these heavy fuels oil n°6 is very different from other petroleum products like the "Arabian Light" crude oil, a diesel oil, or a gasoline which are respectively biodegradable up to 45%, 90% and 100% (Ballerini et al. 1979, Quinn J.W. 1995, Solano-Serena et al. 2000). But even though the biodegradation level is limited, it can be assumed that the biological part of the natural attenuation will contribute to transform the emulsified oil in an inert bituminous residue.

The results presented in this work indicate that the clean-up strategy should consist in removing mechanically as much oil as possible from the sea and the coast. Because of a limited biodegradability of the fuel biological treatments can not be considered for remediation. Physical and physico-chemical treatments like washing, incineration, thermal desorption… must be the most effective technologies to clean up the material removed from the shore.

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