

The Erika Oil Spill, 10 Years After: Assessment of the Natural Weathering of the Oil and Natural Recovery of Vegetation

March 2, 2011

Ronan Jézéquel, Florence Poncet

Cedre

715, rue Alain Colas – BP 20413

29218 BREST Cedex

FRANCE

ronan.jezequel@cedre.fr

florence.poncet@cedre.fr

ABSTRACT

On December the 12th, 1999, the tanker Erika broke up and sank off the coast of Brittany (France) leading to the spill of 20,000 tons of a heavy fuel oil. 400 km of the French Atlantic coastlines were polluted. Because of the characteristics of the oil (a very heavy fuel oil with a high content of heavy chemical compounds) and the severe weather conditions (a centennial storm with spring tides) when the oil came on shores, the Erika spill was one of the most severe accidental release of oil along the French coastlines, all types of habitat were concerned, pollution reached the supratidal zone affecting terrestrial vegetation and lichens.

During five years after the spillage, in the frame of a monitoring program on Erika oil spill consequences, initiated by the French Ministry of Environment, Cedre was in charge of a study on the natural degradation of oil remaining in different locations selected according to their environmental conditions (solar exposure, wave action, ...) and of second one, on the natural recovery of littoral plant communities in sixty locations in which a hundred plots representing main vegetation types, various degrees of oiling and treatments applied were established. Vegetation phytosociological indices, monitored during five years, suggest moderate short or medium term impact even if oil persistence was observed. At the end of monitoring program, impact is still observable on slow growing communities (lichens) or where clean-up has induced severe degradation.

Ten years after the wreckage, locations selected were revisited: vegetation indices were examined (species composition, vegetation coverage) and samples of oil were collected for analyses.

Samples of weathered oil were collected in triplicate, extracted with methylene chloride and then purified through alumina-silica microcolumn. After extraction, GC-MS/MS analyses were performed in order to assess the degradation of alkanes and PAH compounds.

INTRODUCTION

When released into the environment, petroleum is subject to a variety of natural processes such as evaporation, dissolution, dispersion, biodegradation and photo-oxidation (National Research Council, 2002). The intensity of these natural degradation processes depend on the environmental conditions such as solar radiation, wave exposure, temperature, tidal level, substrate nature, ... Consequently, according to the site location and the environmental conditions in which the oil persists, the weathering rate of the pollutant could be significantly different.

The Erika oil spill along the French coastline was severe because of the nature of the heavy fuel oil and the unusual weather conditions (centennial storm) during the spillage combined with the extreme tides of the year. Some oil projections reached 20 meters high in exposed cliffs. The contamination was scattered along approximately 400 km of the west coastline of France, and most of the oil deposits occurred on the upper part of the intertidal zone up to the supratidal zone. Intertidal salt-marshes, beaches and dunes as well as rocky phanerogam and lichen communities were oiled. In vegetated areas, initial oil emulsion deposits ranged from a few millimetres to 5 centimetres thick on sloping areas or cliffs and reached 15 to 20 cm on flat areas such as intertidal marshes. A major clean-up operation was led under the French Polmar national organization and lasted 2.5 years. As a considerable amount of information on the oil's impact on vegetation (chemical toxicity or physical effect of coating) and on the impact of clean-up had been published since the seventies (Baker, 1970 and 1971; Bretau, Levasseur, 1983; Baker et al., 1993, Sell et al., 1995), specific technical recommendations for vegetation clean-up and protection of adjacent areas and accesses were rapidly set up. To avoid major, long term impact, the removal of thick accumulations was a priority; however vegetation clean-up was not systematic when it was medium to lightly oiled, and even a few small spots remained uncleaned for scientific interest. An extensive five year environmental assessment program was initiated by the French Ministry of the Environment; *Cedre* was in charge of two studies, one on the natural degradation of oil remaining in different locations selected according to their environmental conditions (solar exposure, wave action...) and a second, on the natural recovery of coastal plant communities.

For the vegetation study, 175 permanent quadrats were set up in July 2000 on 60 sites, representative of the main vegetation types, various degrees of oiling and treatments applied but also of incidental damage due to response actions. The parameters monitored were species composition, ground cover, stem height and phenology. Photos completed these data and were used to make a qualitative comparison of vegetation aspect through years.

The study showed: (i) minimized impact on heavily oiled vegetation (e.g. salt marshes) due to adequate removal of the bulk of the oil before the regrowth of plants in the spring, limiting mortalities due to coating; (ii) light to moderate short (1 year) and medium term (2-3 years) damages due to oil or oil and clean-up consisting in the reduction of vegetation cover due to mortalities, tar crust persistence or plant destruction due to the manual scraping of oil deposits; (iii) only little evolution of the vegetation composition was observed in a few quadrats. By the end of the program in 2005, in most quadrats monitored, the vegetation composition and cover were comparable to adjacent vegetation, even when residual oil was still persistent. Long term impact (>5 years) was observed on slow-growing communities like lichen, on grey (fixed) dunes or on vegetation damaged by clean-up operations as on some heavily oiled rocky areas cleaned by high pressure hot water washing (HPHW). These results were consistent with previous oil spill impact studies (Levasseur and Jory, 1982; Moore et al, 1998; Michel et al 2005). At this time, residual oil persistence was noted in the supratidal zone, such as a dry film or pavement on shingles, upper beaches, rocky shores and cliffs and slightly sticky in some dripping vegetated cliff crevices. In the intertidal zone, a residual sticky layer remained under the dry tar crust in some uncleaned areas in saltmarshes (low hydrodynamic conditions).

Ten years after the wreckage, selected locations were revisited, vegetation indices were examined (species composition, vegetation coverage) and samples of oil were collected for analysis. Sites were chosen among the few where vegetation was voluntarily left partially or totally uncleaned to avoid incidental impact of operations as vegetation was developing normally

the spring after the spill. Two sites (6701, C1/C2) were left uncleaned for scientific observation due to their difficult accessibility by the public. The oil sampling site C1 and C2 represented the main accumulation left for evolution monitoring on the affected coast.

METHODS & MATERIALS

Site location and sampling references are presented in Table 1 and Figure 1.

Vegetation survey

Selected sites were surveyed in September 2010. Species composition, vegetation coverage and phenology were compared to 2005 data.

Oil sample extraction

Approximately 2g of samples were extracted with 30 mL methylene chloride at 100°C and 2000 psi for 14 minutes using a Dionex ASE 350 accelerated solvent extractor. Extractions of sediment were performed twice for each sample. Organic extracts were dried over Na₂SO₄ (activated at 400°C for 4 hours) and concentrated to approximately 2 mL using a Syncore (Büchi Labortechnik AG, Flawil, Switzerland). Approximately 20 mg of extracts were then fractionated using SPE (Solid Phase Extraction) cartridges (silica/cyanopropyl (SiO₂/C3-CN) (1.0/0.5 g, 6 ml) (Interchim, Montluçon, France)) (Alzaga *et al.*, 2004): saturate and aromatic fractions were eluted simultaneously with 8 mL of methylene chloride / pentane mixture (80/20, v/v) and concentrated to approximately 2 mL under a stream of nitrogen.

Sample analysis

The analyses were performed by Gas Chromatography coupled to a Mass Spectrometry (GC-MS/MS). The GC was an Agilent 7890A (Agilent Technologies, Little Falls, DE, USA) coupled to an Agilent 7000 Triple Quad Mass Spectrometer. Samples of 1 µl were injected automatically with an Agilent 7693 autosampler. The injector temperature was maintained at 300°C and the interface temperature was 300°C. The GC temperature gradient was: from 50°C (1min) then 10°C/min to 150°C and 5°C/min to 320°C (10 min). The carrier gas was Helium at a constant flow of 1 mL/min. Two capillary columns HP 5 MS UI (15 m x 0.25 mm ID x 0.25 µm film thickness) were connected through a splitter system used to perform a 3 minutes backflush of the first column at the end of each run. The triple quad system was operated in Single Ion Monitoring (SIM) with a minimum of 1.5 cycles/s. Alkanes and PAHs were identified by comparison with appropriate external standards. Compounds not present in the standard mixture were identified on the basis of retention times in published data (Daling and Faksness, 2002).

RESULTS & DISCUSSIONS

Impact of oil on vegetation

The Results of observations of each selected quadrat are presented in Table 2. Most of the sampled areas are located at supratidal level, seldom reached by wave action. In this upper level, sticky oil remained on oil sampling sites C1/C2, on which an accumulations of 10 cm thick was trapped among blocks and boulders in the supratidal zone. Apparently, since the extreme conditions prevailing in 2009 when the oil was thrown oil very high up, these boulders were not moved again and oil remained trapped since its deposit.

The oily dry film (thickness 1 mm on average) still present in 2005 on exposed surface of rock in quadrat n°4904, was no longer visible in 2010, but no lichen colonization was yet observed at the place of the oil stain. For all vegetation quadrats where the thickness of oil still observed in 2005 was greater, such as tar crusts of 1-3 cm on average, these tar crusts were slowly disappearing but residues were still present in 2010.

In intertidal zones, sea erosion of muddy marsh fringes has accelerated the disappearance of part of this pavement (quadrat 1101, 6104) but the tar layer remained where erosion had not been effective. On rocky platforms, piece of rock knocked down by waves also accelerated the natural removal of tar (quadrat 0401).

Since 2005, no significant impact on vegetation development and species composition due to this tar pavement residue was noticeable.

Three sites were monitored for indirect impact due to high pressure/hot water washing. In two sites (quadrat 1204 and 4707) where vegetation and surface soil had been destroyed, recovery is far from complete, but recolonization by crustose lichen (*Lecanora atra*) and *Ramalina siliquosa* had significantly increased (from 40 to 80% rock coverage) in the third one (quadrat 4904).

Oil weathering

Analysis of the original Erika oil

$\alpha(H), 21\beta(H)$ -hopane ($m/z=191$) (30 ab) was used as internal conservative biomarker during analyses (Prince *et al.*, 1994, Venosa *et al.*, 1997). Consequently, the distribution of *n*-alkanes and PAHs are presented in Hopane Unit. Figure 2 highlights the preservation of hopane compounds between the original sample of Erika oil and some of the samples collected after ten years of weathering.

The hopanes family was systematically studied for each sample in order to fingerprint the oil. Among all of the collected samples, two were attributed to other products due to differences between source ratios (Daling and Faksness, 2002) and also due to the absence of 30d (diahopane), a compound specifically found in the Erika oil.

The results of the GCMS analysis of the original Erika oil are presented in Figures 3 and 4. The alkanes were distributed between *n*-C₁₁ and *n*-C₄₀ and centered close to *n*-C₂₄. This difference of distribution is common in Heavy Fuel Oil (HFO) constituted by a mixture of a distillate (LCO - light cycle oil in this case) with a residue (Vacuum Distillate Residue in this case) (Guyomarch *et al.*, 2001), in order to fluidize it and achieve HFO commercial requirements (mainly viscosity) (Lewis, 2002). In the same way, PAHs distribution is characterized by the abundance of Naphthalene compounds (40% of the total PAHs), which is typical of LCO addition.

Analysis of the weathered samples of Erika oil

The percentages of depletion of *n*-alkanes and PAHs are presented in Figure 5. Alkanes and PAHs are classified according to their molecular weight. For *n*-alkanes, low molecular weight (LMW-Al) corresponds to the sum of *n*-C₁₁ to *n*-C₂₄ and heavy molecular weight (HMW-Al) to the sum of *n*-C₂₅ to *n*-C₄₀. For the PAHs, low molecular weight (LMW-PAH) includes Benzo[thiophenes], Naphthalenes, Biphenyl, Acenaphthene and Fluorenes while the heavy PAHs (HMW-PAHs) include Phenanthrenes, Anthracenes, Dibenzothiophenes, Fluoranthenes, Pyrenes, Chrysenes, Benzo[a]anthracene, Benzo[b]fluoranthene, Benzo[k]fluoranthene,

Benzo[e]pyrene, Benzo[a]pyrene, Perylene, Indeno(1,2,3-cd)pyrene, Dibenz(a,h)anthracene and Benzo(g,h,i)perylene.

After 10 years of weathering, most of the samples (except C2 and D2) present a degree of degradation greater than 90% for *n*-alkanes (LMW and HMW) and LMW-PAHs. All these compounds are known to be sensitive to natural degradation processes, such as dissolution, evaporation and biodegradation for the lightest compounds, and biodegradation for the heaviest *n*-alkanes (Prince, 1998). For the HMW-PAHs, degradation rate was between 43% (C2) and a maximum of 87% (E1).

A principal component analysis (The Unscrambler v9.7, CAMO software AS) was employed as a means of easily isolating different groups of samples according to the degradation rate of *n*-alkanes and PAHs (Figure 6). The PCA results clearly distinguish three groups of data: the first one includes samples A2, A4, B2, D1, E1 and E2; the second includes A1, A3, B1, C1 and the last group includes samples C2 and D2. Referring to Table 1, these groups correspond exactly to exposure conditions (except for E2): Group I for the highest level of exposure, Group II for a moderate exposure, and Group III for the sheltered samples. Concerning E2, as the oil was found under 5 cm of mud, this sample was classified as a sheltered one, nevertheless, this type of substrate does not ensure that the oil, found in the form of tarball, was never exposed and consequently, the heaviest PAHs had degraded (82%) as observed for the exposed samples.

These results show that the differences in degradation rates of Erika oil were mainly due to the exposure level and consequently could probably be attributed to the photo-oxidation process. To confirm this, losses of chrysenes (parent and alkylated forms) were calculated for the previously defined groups and are presented in Figure 7. Chrysenes, as with all heavy polycyclic compounds, are known to be sensitive to photo-oxidation process and not easily biodegraded (Wang *et al.*, 1994, Garrett *et al.*, 1998, Hadibarata, 2009), this is indeed what Figure 7 demonstrates: chrysenes degradation is significantly different between each Group ($63 \pm 4\%$ in Group I, $28 \pm 8\%$ in Group II and $5 \pm 2\%$ in Group III). Nevertheless, considering each Group individually, the degradation rate are inversely proportional to the degree of alkylation which is typical of the biodegradation process ($C > C1 > C2 > C3$) (Wang *et al.*, 1994, 1997, Michel *et al.*, 1999) suggesting during ten years in environment, heaviest compounds are subject to biodegradation and photo-oxidation.

CONCLUSIONS

Ten years after the release of 20,000 tonnes of a heavy fuel oil on the French Atlantic coastline, samples of weathered oil were collected in order to study their degradation based on *n*-alkanes and PAHs depletion. Most of the samples of oil collected in exposed or moderately exposed conditions present a heavy weathering through the total loss of the lighter-weight compounds and a significant degradation of the heaviest compounds. Two samples collected in sheltered condition (under a cobble for C2 sample and trapped between blocks for D2 sample) were the least degraded even if the degradation rate of *n*-alkanes and PAHs exceed respectively 78% and 66%, which can be considered as a high level of degradation.

In term of impact on vegetation, the least degraded sample of oil collected (C2) was collected in a block and cobble accumulation where no vegetation was growing. For the other oil samples, no significant impact of these pavement residues on species composition or vegetation development and coverage was observed excepted in lichen communities. Tar stain (oil dry film) on medium exposed rocks covered by lichen communities have disappeared since 2005, but are

not yet replaced by new growing lichen. The oil's long persistence is noticeable in supratidal zones.

On intertidal zones, oil residues remain only in habitat with low stable hydrodynamic conditions, like salt marshes. Locally in marshes surveyed, some marine erosion of the fringe of marsh sediment is observed and contributes to oil disappearance.

Two quadrats intensively cleaned-up with HPHW washing show poor soil and vegetation recuperation ten years after the spill.

ACKNOWLEDGEMENTS

During the study, a significant part of the chemical analyses were performed in order to fingerprint the sample of oil collected in the environment. This work, undertaken with the support of Total Company and in collaboration with CReG laboratory (Total Gonfreville – France), will be presented during the next CEN meeting (European Committee for Standardization) and will continue over a specific study dedicated to the fingerprint of very weathered oil.

REFERENCES

- Alzaga, R., Montuori, P., Ortiz, L., Bayona, JM. and J. Albaiges. 2004. Fast solid-phase extraction-gas chromatography-mass spectrometry procedure for oil fingerprinting. Application to the Prestige oil spill. *Journal of Chromatography A*. 1025, Issue 1, 30:133-138.
- Daling, P.S., L.G. Faksness. 2002. Laboratory and reporting instructions for the CEN/BT/TF 120 Oil Spill Identification-Round Robin Test-May, 2001. Technical report, SINTEF report STF66 A02027.
- Baker, J.M., 1970. The Effects of Oils on Plants, *Environmental Pollution*, 1:27-44.
- Baker, J.M., 1971. Seasonal effects. In : *The Ecological Effects of Oil Pollution on Littoral Communities*, Ed. E.B. Cowell, pp.44-51. Institute of Petroleum, London.
- Baker, J.M., L. Guzman, P.D. Bartlet, D.I. Little, and M. Wilson, 1993. Long-term Fate and Effects of Untreated Thick Oil Deposits on Salt Marshes, In *Proceedings of the 1993 International Oil Spill Conference*, American Petroleum Institute, Washington DC, pp. 395- 399.
- Bretau, F., Levasseur, J. E., 1983. Généralités sur les conséquences d'une pollution marine de marais intertidaux par hydrocarbures : impact sur la couverture végétale et l'environnement. Rapport final contrat 82/132, *Cedre/Université de Rennes 1*, 75p.
- Garrett, R.M., Pickering, I.J., Haith, C.E. and R.C., Prince. 1998. Photooxidation of crude oils. *Environmental Science and Technology*. 32, 3719– 3723.
- Guyomarch, J. Budzinski, H., Chaumery, C., Haeseler, F. Mazeas, L., Merlin, FX and J. Oudot, 2001. The ERIKA Oil Spill: Laboratory Studies carried out to Assist Responders. *Proceedings of the 2001 International Oil Spill Conference*, American Petroleum Institute, Washington, D.C., 637-647.

Hadibarata, T., Tachibana, S. and K. Itoh. 2009. Biodegradation of chrysene, an aromatic hydrocarbon by *Polyporus* sp. S133 in liquid medium. *Journal of Hazardous Materials*. 164. 911-917.

Levasseur J., et Jory M.L., 1982. Rétablissement naturel d'une végétation de marais maritimes altérée par les hydrocarbures de l'*Amoco Cadiz* : modalités et tendances. *In* : Ecological Study Of The Amoco Cadiz Oil Spill, Report of the NOAA-CNEXO Joint Scientific Department, Oct. 1982 : 329-362 pp.

Lewis, A. 2002. Composition, properties and classification of heavy fuel oils. Proceedings of the Third R&D forum on High Density Oil Spill Response. Brest. France. International Maritime Organization. London. 39-45.

Michel, J. and M.O. Hayes. 1999. Weathering Patterns of Oil Residues Eight Years after the Exxon Valdez Oil Spill. *Marine Pollution Bulletin*, Vol. 38, 10, 855-863.

Michel, J., Hayes M. O., Getter C. D., Cotsapas L., 2005, The gulf war oil spill twelve years later : consequences of ecoterrorism. Proceedings of the 2005 International Oil Spill Conference, American Petroleum Institute, Washington, DC, Publ.14718 A (CD-ROM).

Moore, J.J., Little, A.E., et Dyrinda, P.E.J., 1998. *Sea Empress* oil spill : the ecological effects of shoreline clean-up. *In* « The *Sea Empress* oil spill ». Proceedings of the conference held in Cardiff on 11-13 February 1998, edited by Edwards R. & Sime H. : 475-482.

National Research Council. 2002. Oil in the Sea III: Inputs, Fates and Effects, National Academy of Sciences, Washington DC.

Prince, R. C., Elmendorf, D. L., Lute, J. R., Hsu, C. S., Haith, C. E., Senius, J. D., Dechert, G. J., Douglas, G. S. and E. L. Butler. 1994. 17 α (H),21 β (H)-hopane as a conserved internal marker for estimating the biodegradation of crude oil. *Environmental Science and Technology*. 28, 142-145.

Prince, R.C., 1998. Crude oil biodegradation. *The Encyclopedia of Environmental Analysis and Remediation*. Wiley, New York. Vol. 2. 1327-1342.

Sell, D., Conway, L., Clark, T., Picken, G.B., Baker, J. M., Dunnet, G.M., McIntry, A. D., and Clark, R. B., 1995. Scientific criteria to optimize oil spill cleanup. Proceedings of the 1995 Oil Spill Conference. American Petroleum Institute, Washington DC, pp 595-610.

Venosa, A.D., Suidan, M.T., King, D., and Wrenn, B.A. 1997. Use of hopane as a conservative biomarker for monitoring the bioremediation effectiveness of crude oil contaminationg a sandy beach. *Journal of Industrial Microbiology and Biotechnology* 18:131-139.

Wang, Z., Fingas, M. and G., Sergy. 1994. Study of 22-year-old Arrow spill sample using biomarker compounds by GC/MS. *Environmental Science and Technology*. 28. 1733-1746.

Wang, Z., and M., Fingas. 1997. Developments in the analysis of petroleum hydrocarbons in oils, petroleum products and oil-spill-related environmental samples by gas chromatography. *Journal of Chromatography A*, 774, 51-78.

2011 INTERNATIONAL OIL SPILL CONFERENCE

Site	Oil Sample number	Veget quadrat ref.	Description and observations	Exposure ⁽¹⁾
Kerliguit		0401	Brackish marsh (<i>Phragmites australis</i> , <i>Glaux maritima</i> , <i>Juncus maritimus</i>) on rocky platform with organic soil and freshwater seepage. Veget. uncleaned; initial oiling > 1cm. Hard tar at the foot of some of the plants. Supratidal level.	-
Porz Teg	A 1		Small open rocky bay. Soft asphalt pavement between rock crevices. Upper intertidal level.	+
	A 2	1101	Brackish marsh on peat and clay platform upper part of the bay (<i>Scirpus maritimus</i> ie. <i>Bolboschoenus maritimus</i>). Veget. uncleaned; initial oiling > 1cm. Hard tar at the foot of plants. Upper intertidal level.	++
	A 3		Pebble beach at the foot of the peat marsh. Soft oil on the unexposed side of a pebble. Upper intertidal level.	+
	A 4		Hard asphalt on the exposed side of a pebble. Upper intertidal level.	++
Porz Lamat	B1		Small open rocky bay. Asphalt patches on rocks. Upper intertidal level.	+
	B2	1204	Grass with <i>Elymus sp.</i> and <i>Festuca rubra</i> on rocky platform. HPHW on adjacent rock having impacted veget. and soil. Hard oil tar at the foot of plant. Supratidal level.	++
Port Pilote		6701	Rocky crevice with <i>Crithmum maritimum</i> . Initial oiling < 1cm, uncleaned for scientific interest. Hard oil tar at the foot of plant. Supratidal level.	-
Les Roches Grises		2202	Exposed shingle beach on upper part of a rocky platform with <i>Elymus sp.</i> Veget. uncleaned; initial oiling > 1cm. Hard tar at the foot of plant. Supratidal level.	++
		2201	Exposed shingle beach on upper part of a rocky platform with <i>Crithmum maritimum</i> . Veget. uncleaned; initial oiling > 1cm. Hard tar among shingles.	++

			Supratidal level.	
Crique Décharge	D1		Exposed rocky platform with <i>Crithmum maritimum</i> . Uncleaned. Hard asphalt on the exposed rock. Supratidal level.	+
	D2		Accumulation of soft oil in a rock crevice. Presence of <i>Crithmum maritimum</i> . Supratidal level.	-
Crique Géodésique	C1		Exposed creek on a rocky cliff coast, with an accumulation of blocks and boulders at the foot of the cliff. Uncleaned for scientific interest. Asphalt pavement on the surface of a block. No soil, nor vegetation. Supratidal level.	++
	C2		Accumulation of soft oil trapped between blocks and boulders. No soil, nor vegetation. Supratidal level.	-
La Grande Butte		4707	Rocky cliff edge platform where chasmophyte community disappeared after intensive high pressure washing on heavily oiled platform. Supratidal level.	++
Port aux Rocs		4904	Exposed rocky platform with lichen community. Residual tar film stains and rock partially cleaned by HWHPwashing. Supratidal level.	+
La Parracaud	E1	6104	<i>Spartina maritima</i> salt marsh in a wide open bay. Veget. uncleaned; initial oiling > 1cm. Hard asphalt pavement at the surface of mud. Intertidal level.	++
	E2		Hard tar ball under 5 cm of mud. Intertidal level.	-

Table 1. Site location of vegetation quadrats and corresponding oil samples references. Main characteristics of vegetation, habitat conditions, degree of oiling and clean-up technique applied. (*): as most of the selected areas are located at supratidal level, “exposure” is referring mainly to solar radiation rather than wave action.

Site name	Sample n°	Ref quadrat	2005 results	2010 results
Kerliguit		0401	<i>Phragmites australis</i> , <i>Glaux maritima</i> , <i>Juncus maritimus</i> ... No impact on species comp. or veget. coverage (95%). Hard tar at the foot of some of the plants slowly decreasing.	Species composition idem. Vegetation coverage idem. Dry tar residues.
Porz Teg	A 2	1101	<i>Scirpus maritimus</i> (<i>Bolboschoenus maritimus</i>). Erosion of the micro-cliff. No impact on species comp. Hard tar at the foot of plants.	Species composition idem. Progress of erosion. Dry tar residues.
Porz Lamat	B2	1204	Grass with <i>Elymus sp.</i> and <i>Festuca rubra</i> on rocky platform Erosion due to clean-up(HPHW) Hard tar at the foot of plant.	Species composition idem. No progression of veget. Coverage. Dry tar residues.
Port Pilote		6701	Crevice with <i>Crithmum maritimum</i> . Uncleaned for scientific interest. No impact on plant development Hard tar at the foot of plant.	More species. Good development of plant. Dry tar residues.
Les Roches Grises		2202	Shingle beach with <i>Elymus sp.</i> No impact on species comp. Vegetation cover (80%) due to hard tar pavement.	Species composition idem. Vegetation coverage idem. Dry tar residues.
		2201	<i>Crithmum maritimum</i> . No impact on species comp. No impact on veget. Cover. Hard tar among shingles.	Species composition idem. Vegetation coverage idem. Dry tar residues.
La Grande Butte		4707	Rocky cliff edge platform. Veget. disappeared after intensive high pressure washing. Only 4 species back (average of 10 in references). Veget. cover 4%.	Species comp. idem. No significant progress of veget. cover.
Port aux Rocs		4904	Rocky platform with lichen. Unclean rock with residual tar	Stain disappeared, but no colonization by lichen yet.

			film stains and no lichen. Rock washed by HWHP: 40% lichen recolonization started.	Washed area: colonization of up to 80% of rock surface.
La Parracaud	E1	6104	Monospecific <i>Spartina maritima</i> saltmarsh. Uncleaned for scientific interest (oil deposit 2-5 cm thick). Decreasing vegetation cover due to erosion (96% to 85%) No impact on species comp. and development. Hard asphalt pavement on the surface of mud.	Progress of erosion of the fringe of the marsh, quadrat partially disappeared. Few residual tar pavement fragments still present. Development of plants and veget. coverage similar to adjacent areas.
	E2		Hard tar ball under 5 cm of mud.	Idem E1.

Table 2. Results of vegetation assessment in quadrat followed since 2000: comparison between 2005 and 2010 observations on species composition, vegetation coverage, habitat characteristics and presence of oil residues.

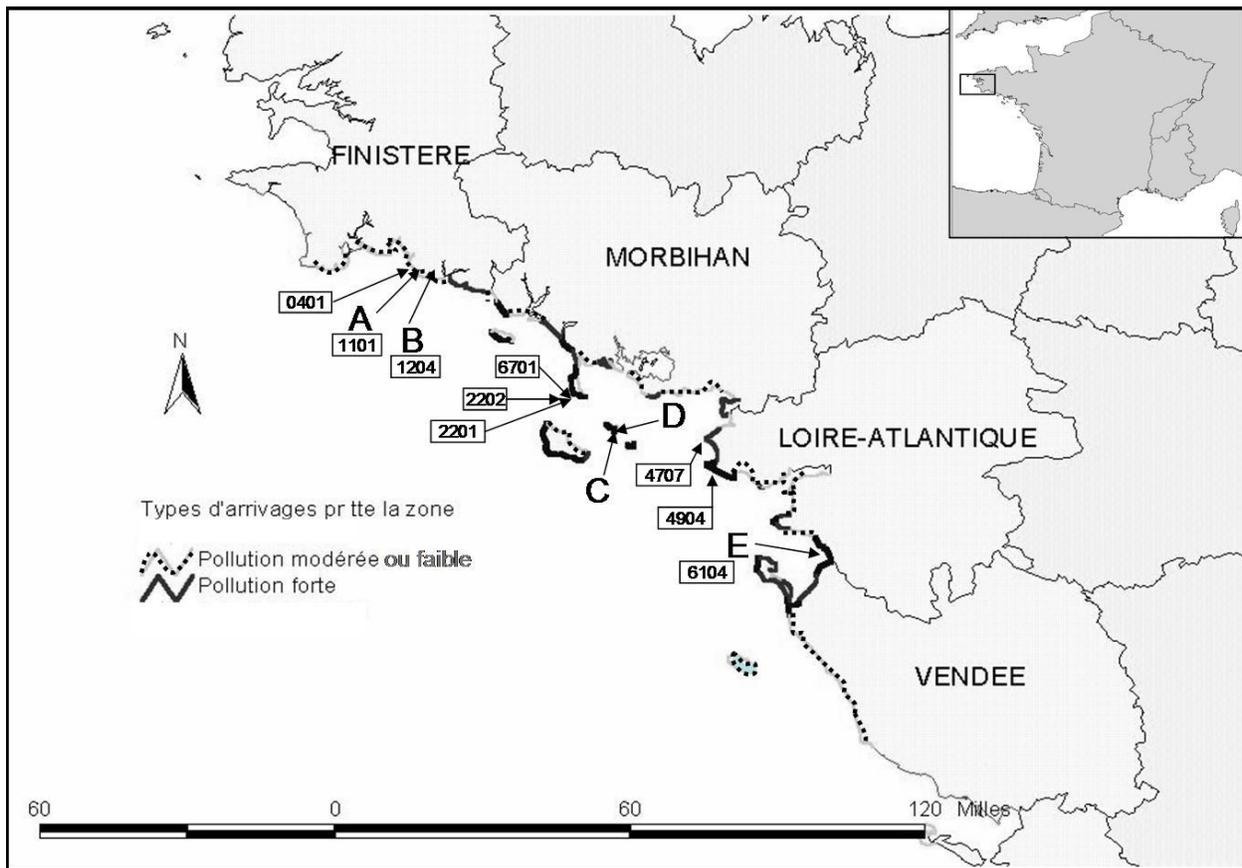


Figure 1. Vegetation quadrat and sampling site location map

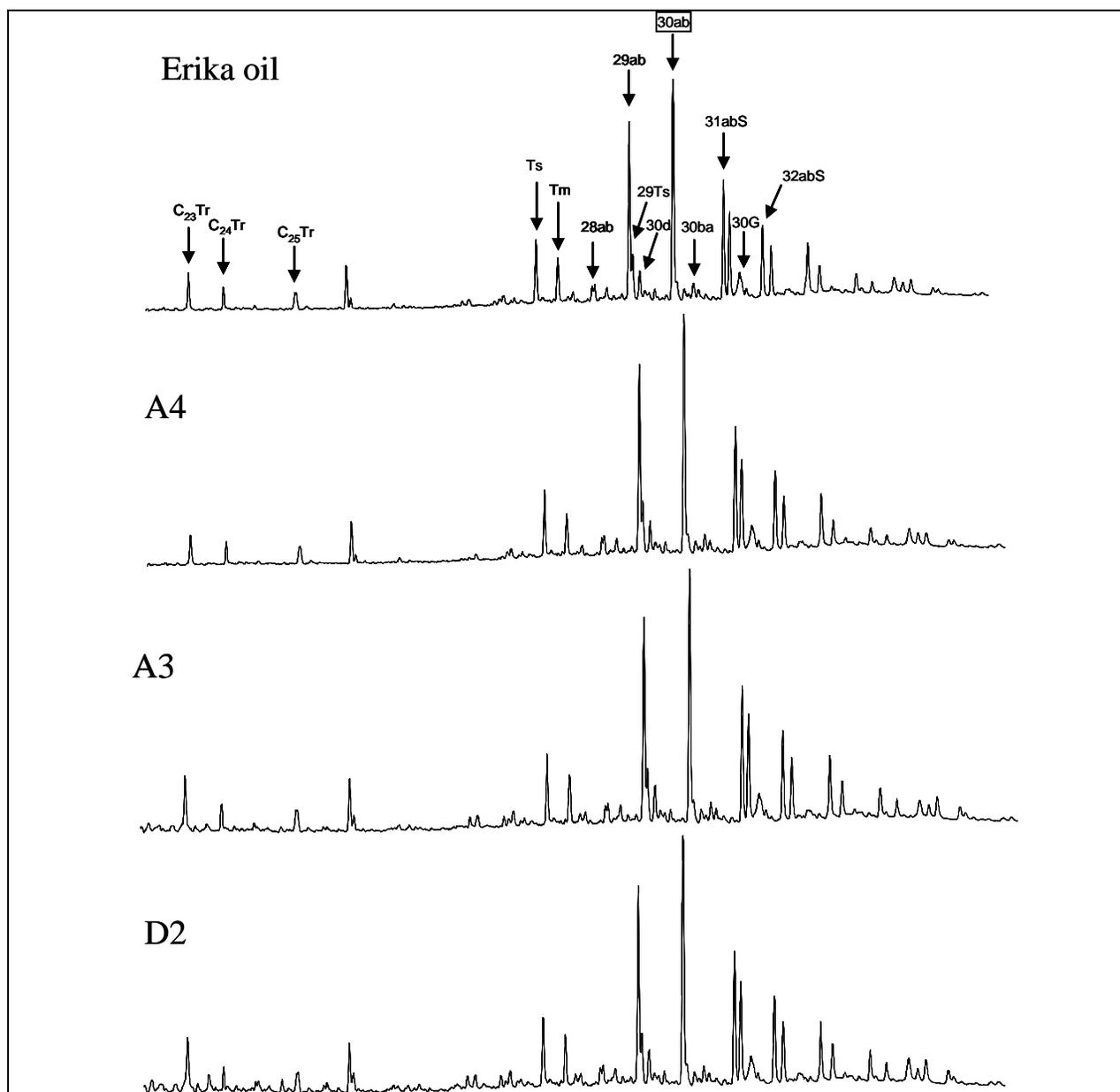


Figure 2. GC-MS chromatogram illustrating the preservation of biomarker terpanes (m/z 191) between the original Erika oil and samples (A4, A3 and D2) collected 10 years after the oil spill. (30ab was used as an internal standard).

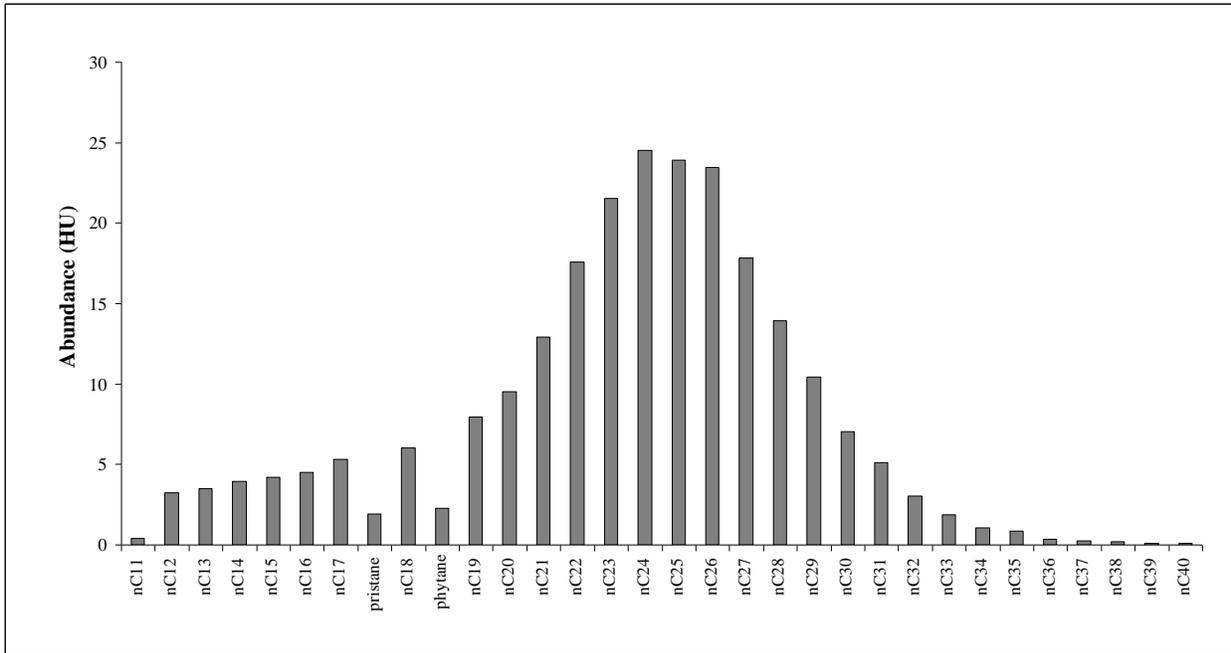


Figure 3. *n*-alkanes distribution (Hopane Unit) for the original Erika oil.

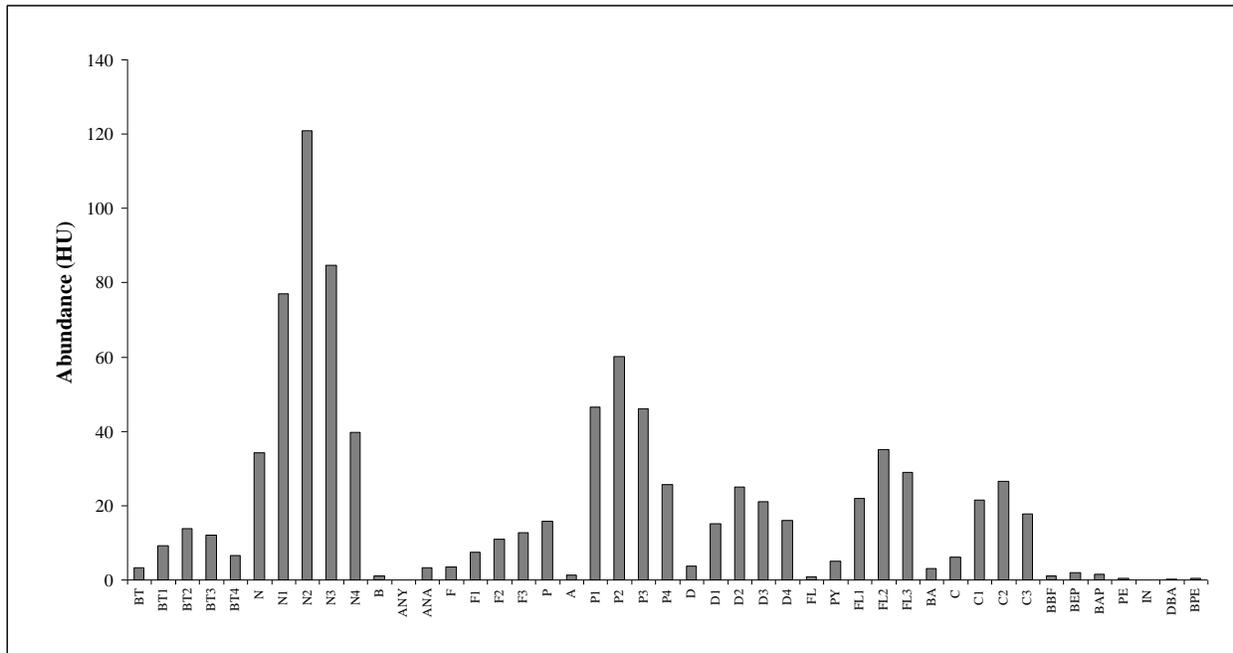


Figure 4. PAHs distribution (Hopane Unit) for the original Erika oil.

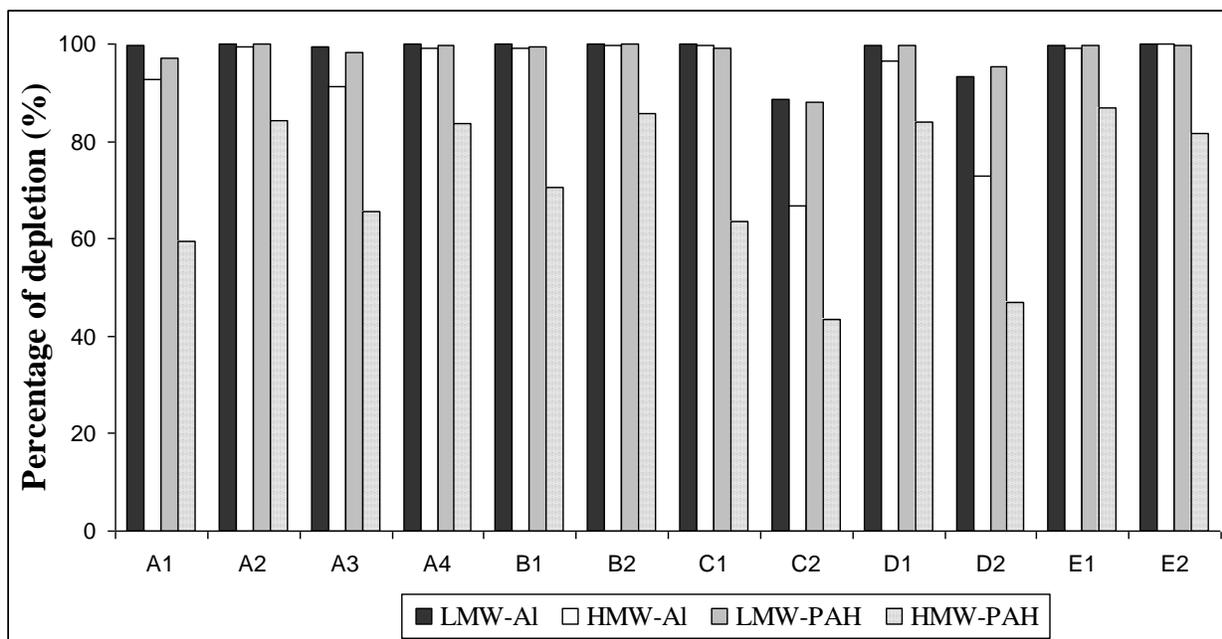


Figure 5. Degradation of low and high molecular weight *n*-alkanes (LMW-Al and HMW-Al) and low and high molecular weight PAHs (LMW-PAH and HMW-PAH) for samples of oil collected ten years after the Erika spill at different site locations (site A, B, C, D and E).

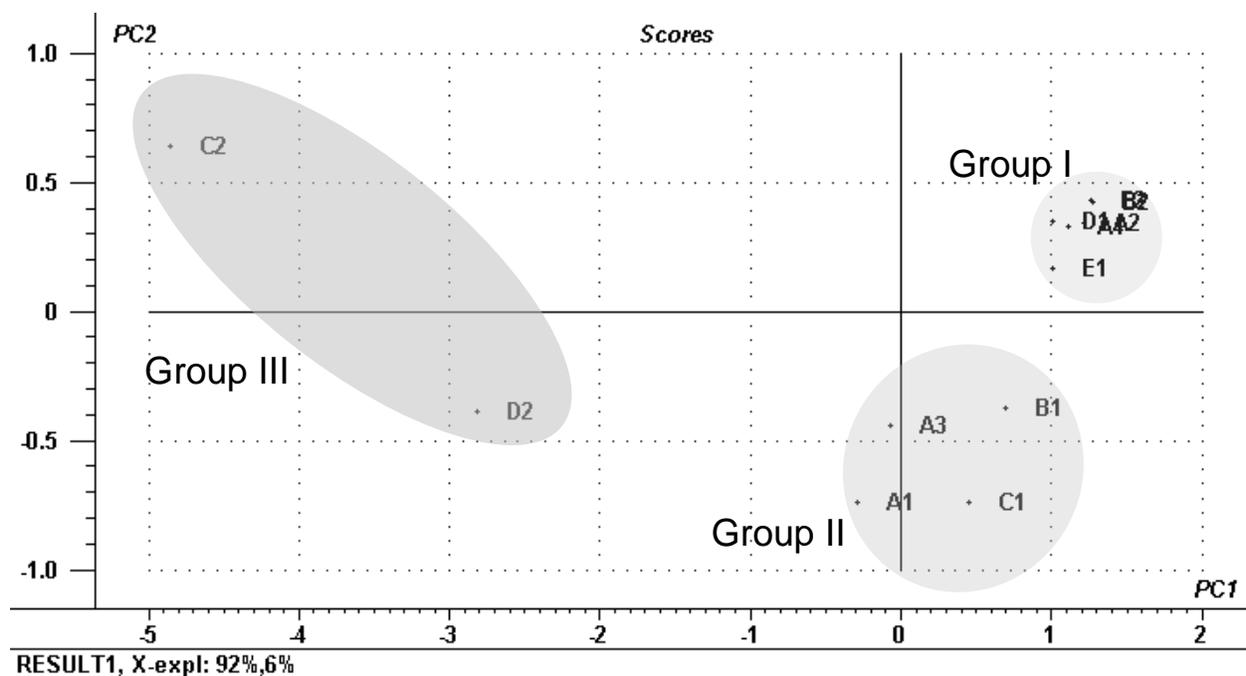


Figure 6. Principal Component Analysis (PCA) (scores plot) for *n*-alkanes and PAHs degradation. (98% of the total variance of *n*-alkanes and PAHs degradation is explained with PC1 and PC2). The reference of each plot indicates the sample numbers.

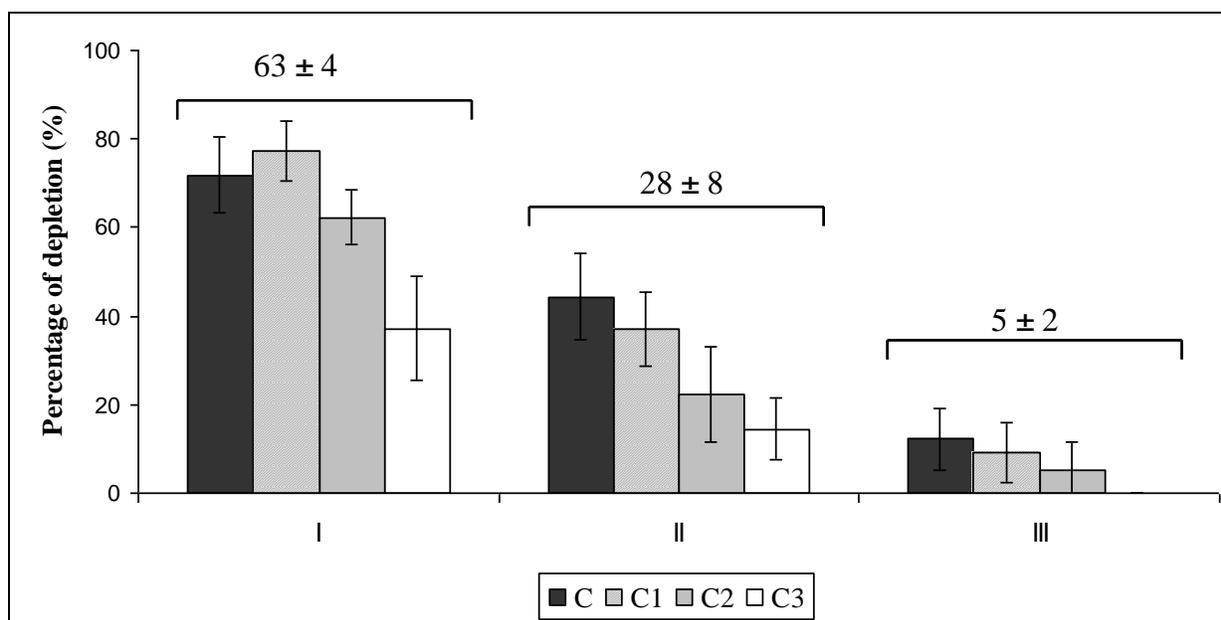


Figure 7. Degradation of chrysene (C) and some of its alkylated derivatives (C1, C2 and C3 refer to methyl-, dimethyl- and ethyl-, and trimethyl-, methyl-ethyl and propyl-chrysenes, respectively) in oil samples from Groups I, II and III. The number above the brackets represents the mean and standard deviation of percentage of degradation of total chrysenes (C + C1 + C2 + C3).