CONTAMINATION CHIMIQUE DES MOLLUSQUES INTERTIDAUX PAR LES HYDROCARBURES AROMATIQUES POLYCYCLIQUES SUITE AU NAUFRAGE DE L'ERIKA : PREMIER BILAN
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ABSTRACT

On December 12th 1999, the Erika tanker was wrecked off the coast of Morbihan, spilling an important quantity of fuel oil and causing the contamination of more than 400 kilometres of coastline from south of Finistère to Vendée. The fuel oil of the tanks of the ship (fuel oil no6) contained aromatic compounds, including polycyclic aromatic hydrocarbons –PAH-, heterocyclic sulfur compounds, and their alkyl derivatives.

The levels of contamination by PAH and by their alkyl derivatives were studied in the samples of different species of intertidal mollusks taken monthly from December 1999. The sampled species are mainly feeders like mussels (Mytilus edulis), and oysters (Crassostrea gigas) or detritus-feeding animals like cockles (Cerastoderma edule). Mollusks are used like biological indicators of the contamination of coastal environment and make it possible to follow the temporal evolution of this contamination since the wreck. The sites of sampling spread from Finistère to Vendée make it possible to specify the spatial extension of the contamination.

The analyses were carried out on beforehand grounded and lyophilised tissues, by means of a system of automatic and accelerated extraction by hot and pressurized solvent (ASE: Accelerated Solvent Extraction). Next, the organic extracts were purified by gel-permeation chromatography and separated by adsorption chromatography on open silica and alumina column by a gradient of solvents of increasing polarities. The quantitative analyses were done by high-resolution gas chromatography coupled to low-resolution mass spectrometry (ionization by electronic impact); the quantification was done with respect to surrogate internal standards of flooding in selected ions monitoring mode. The compounds monitored as a matter of routine in the samples of mollusks are PAHs, including the 16 PAHs of the EPA list and their alkyl derivatives (methyl-

naphthalenes, methyl-fluorenes, methyl-phenanthrenes/anthracenes, methyl-pyrenes/fluoranthenes, methyl-chrysenes and methyl-benzofluoranthenes), as well as compounds such as sulfured dibenzothiophene, benzonaphthothiophenes and their alkyl derivatives. The quantitative identification of these individual compounds in the samples of mollusks allows to follow the distinctive fingerprint of the contamination by the Erika fuel oil residues.

The concentrations measured in the samples collected just before the arrival of the oil slicks on the coasts constituted the baseline of the chronic contamination in the impacted areas following the wreck. The levels of contamination recorded in the samples, taken each year along the coast as part of the national monitoring network (RNO), provide also data on the reference levels of the contamination in the concerned areas. So, the median concentration, expressed as the sum of 15 PAHs (16 priority PAH of the list EPA except acenaphtylene) and calculated with the results of 4 years of follow-up by the RNO is 146+/-59 ng/g (dry weight) for all the areas concerned by the wreck. This baseline gives an essential reference to determine the return to an initial situation, and to follow the temporal trends of the contamination levels in the mollusks, after the wreck.

The follow-up of the contamination levels in the mollusks shows a very important growth for the days following the arrival of the oil slicks on the coast. The concentrations of the parent compounds (expressed as the sum of PAHs plus the sulfur heterocyclic compounds) and of the alkyl derivatives show very superior levels after the grounding of the oil slicks in comparison with the reference concentrations just before the arrival. For example, for a sample of mussels taken from a site in Loire-Atlantique, these levels are 143 ng/g (dry weight), and 120 ng/g (dry weight) in the sample not concerned by the fuel oil (December 20th 1999); and 2046 ng/g (dry weight) and 10843ng/g (dry weight) after the grounding of the oil slicks.

The temporal variations of the concentrations and the reached maximum levels show differences between the studied sites. So, the maximum concentrations recorded from the end of December 1999 to February 2000 and to May 2000 in Loire-Atlantique, are followed by a reduction of the concentrations in the summer. Rises of concentrations are observed for October and November 2000, resulting probably from a re-mobilization of the fuel oil connected to the hydrodynamic and meteorological conditions of the autumn (storms, high tides). One

year after the wreck, the concentrations of PAH in the mollusks remain superior to the reference levels, particularly in the samples taken in Loire-Atlantique and in Vendée.

The identification of the individual compounds, that define the fingerprint of the contamination, and the distinctive ratios used to characterize this print, also shows unmistakable differences between the references before and after the wreck. The relative distributions of the PAHs in the mollusks are distinguished by the predominance of the alkyl derivatives of the PAH, particularly the methyl-phenanthrenes/anthracenes, methyl-pyrenes/fluoranthenes and methyl-chrysenes. The ratios between the parent compounds and their alkyl derivatives (for example, the ratio phenanthrene/sum of the methyl-phenanthrene, chrysene/sum of the methyl-chrysene) are lower in the samples contaminated by the Erika fuel oil residues than in the reference samples. In the same way, the distinctive indicators, also used to discriminate the oil contaminations from the ones coming from the combustion processes like the ratio fluoranthene/pyrene, show significant differences in the samples contaminated by Erika fuel oil, and also supply indicators to determine the return to a baselevel.

These results obtained from the programme of follow-up and of research on the consequences of the Erika oil spill allow to characterize the impact of an oil spill on the marine mollusks in a coastal environment. They also supply the tools to develop the space expanse and the temporal evolution of the contamination owing to an accidental oil spill.