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Spatial and temporal trends of petroleum hydrocarbons in wild mussels from the Galician coast (NW Spain) affected by the *Prestige* oil spill

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) were determined in tissues of wild mussels (*Mytilus galloprovincialis*) from the Galicia coast (NW Spain) in order to assess the extent of the environmental impact caused by the *Prestige* oil spill (November 13, 2002). Three sampling campaigns were carried out in February, June and November 2003 at 24 stations along the Galicia coast, from La Guardia (Pontevedra) to Ribadeo (Lugo). The spatial distribution of PAHs found in the first sampling period, clearly revealed the central area (Costa da Morte) as the most affected by the oil spill. In these stations, concentrations up to 7780 μg/kg dw of the sum of 13 parent PAHs were found 2–3 months after the spill. Molecular parameters within the aliphatic and aromatic fractions confirmed the presence of the *Prestige* oil in these samples. The levels markedly decreased at most of the stations in the second sampling and recovered to levels found before the spill in November 2003, 1 year after the accident (29–279 μg/kg dw, av. 133 ± 83 μg/kg dw). However, a certain increase was observed in some sites which could be related to the remobilization of oil residues from still unclean intertidal spots or sediments due to the winter marine weather conditions.
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1. Introduction

On November 13, 2002, the *Prestige* tanker, transporting about 77,000 tonnes of heavy fuel-oil, began to leak oil in front of Costa da Morte (Galicia, NW Sapin), and 6 days later, after an erratic towing, broke in two and sank at 240 km off the Northwest coast of Spain (Albaigés et al., 2006). The oil (No. 6 type) had a density of 0.995 kg L^{-1} (at 15 °C) and contained 22% of aliphatic hydrocarbons, 50% of aromatic hydrocarbons and 28% of resins and asphaltenes. The spill affected more than 800 km of the Spanish Atlantic shores and had dramatic ecological and economic consequences for the Galician coastal ecosystem, one of the first producers of mussels worldwide (*Mytilus galloprovincialis*), which should be closed to commercial harvesting.

Soon after the accident a monitoring program was established by the Instituto Español de Oceanografía (IEO) in order to assess the spatial distribution and

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temporal evolution of petrogenic hydrocarbons in the affected area, which involved the sampling of water (González et al., 2006), sediments (Franco et al., 2006) and indigenous populations of mussels.

The use of bivalves as bio-indicators of chemical contamination is widely accepted and is recommended by the International Conventions, such as the OSPAR Commission, the Barcelona Convention and the Helsinki Commission. In this respect, they are widely used in coastal monitoring programmes i.e. the *Mussel Watch* in USA and the *Réséau National d'Observation* (RNO) in France (O'Connor, 1996; RNO, 2002). The interest in using these organisms relies on the fact that they are permanent inhabitants of specific sites and rather resistant to local pollution; they are also filter-feeders and have only a limited metabolic ability. Further, contaminants present in the water column, which may have not been recorded in the sediments, can accumulate in bivalve tissues to levels several orders of magnitude higher than those found in the surrounding seawater (NAS, 1980; Neff, 2002).

In the past, mussels have been successfully used to evaluate the impact of oil spills in coastal habitats, such as the *Exxon Valdez* in the Prince William sound in Alaska (Carls et al., 2001, 2004), the *Aegean Sea* in NW Spain (Porte et al., 2000) and the *Sea Prince* in Korea (Yim et al., 2002) but little information is available on their application to monitoring heavy oil spills (Tronczynski et al., 2004). In this case, it should be taken into account that besides accidental oil spills, hydrocarbons are ubiquitous in the marine environment, so that mussels are chronically exposed to these pollutants, which are linked, among others, to shipping activities (e.g. deballasting waters), urban/industrial runoff and atmospheric deposition. Therefore, the assessment of oil pollution requires the development of analytical techniques that are able to differentiate spill residues from other sources. Characteristic molecular patterns within the aliphatic and aromatic hydrocarbon fractions have been used for this purpose. These include steranes and triterpanes as well as the relative distributions of alkylated polycyclic aromatic hydrocarbons (PAHs) (Bence and Burns, 1995; Porte et al., 2000).

Steranes and triterpanes were suggested as passive tags for source recognition of oil spills by mass fragmentography (Albaigés and Albrecht, 1979). Although these compounds are not known to be toxic to marine organisms, they are useful in marine pollution monitoring as they are quite resistant to degradation and are related to the origin of the oil, so are of diagnostic value for the identification of the hydrocarbon sources. On the other hand, PAHs may be divided in two groups: those with two and three aromatic rings, that in general are only moderately toxic, and those of high-molecular-weight, containing four or more aromatic rings, many of which are mutagenic and teratogenic (Meador et al., 1995). PAHs originated at high temperatures (combustion) are dominated by the parent species, in contrast to crude oils, that contain a wide range of alkyl-derivatives (Youngblood and Blumer, 1975). Mussels are exposed to both types of hydrocarbons, the former preferently associated with the suspended particulate matter and the latter being mainly in the dissolved phase and more bioavailable (Porte et al., 2000).

In order to assess the extent of the impact caused by the *Prestige* oil spill on the Galician coastal ecosystem (NW Spain), using the Mussel Watch concept, three sampling campaigns were carried out in February, June and November 2003 at 24 stations along the coast, from La Guardia (Pontevedra) to Ribadeo (Lugo) (Fig. 1). The samples were analysed for aliphatic and aromatic hydrocarbons to determine the hydrocarbon levels and temporal trends following the accident, and identify the sources. These stations are regularly monitored for PAHs as part of the current IEO monitoring programme, the existence of previous data was important for comparing the contamination levels before and after the spill, and to agree on baseline reference values.

2. Materials and methods

2.1. Sample handling

Wild mussels were collected manually and stored at −20 °C until analysis. At least 50 specimens, of all sizes were pooled as a composite sample representative of each location and sampling period. Once the samples were defrosted, mussels were homogenised and freeze-dried.

2.2. Chemical analyses

2.2.1. Reagents

A PAH mixture containing naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indeno[1,2,3-*cd*]pyrene, dibenzo[*ah*]anthracene and benzo[*ghi*]perylene at 10 mg/L each in cyclohexane was obtained from Dr. Ehrenstorfer (Augsburg, Germany). The following perdeuterated standards, used as surrogates, were obtained from Cambridge Isotope Laboratories (Andover, USA): naphthalene-d_8 (99%), anthracene-d_{10} (98%), pyrene-d_{10} (98%) and benzo[*a*]

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