Marine Pollution Bulletin 62 (2011) 2859–2862

Contents lists available at SciVerse ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Baseline

Polycyclic aromatic hydrocarbons in commercial fish and lobsters from the coastal waters of Madagascar following an oil spill in August 2009

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ARTICLE INFO

Keywords:
Commercial fish
Lobster
Polycyclic aromatic hydrocarbons
Madagascar

ABSTRACT

Concentrations of polycyclic aromatic hydrocarbons were determined in species of commercial fish and lobsters following an oil-spill just off the protected Madagascan coastline. Samples were collected along the coastline within and outside the affected area. Summed PAH concentrations ranged from 1.9 μg kg^{-1} to 63 μg kg^{-1} wet weight, but with no higher molecular weight PAHs (>202 Da) being detected. All concentrations of benzo[*a*]pyrene, benz[*a*]anthracene and dibenz[*a*,*h*]anthracene were <0.1 μg kg^{-1} wet weight, well within the EU and UK set limits for the protection of human health. Additionally, samples were calculated as the benzo[*a*]pyrene toxic equivalency quotient (TEQ) and found to be well below the level of concern in relation to health of human consumers. Evaluation of the biota PAH data indicated the origin of PAH was predominantly petrogenic with >80% arising from oil sources. Profile studies indicate a low-level multisource petrogenic contamination probably representing a pre-spill background for the area.

On 26th August 2009, a cargo vessel grounded off Faux Cap (Cap Sainte Marie) approximately 1.2 km from the southernmost tip of the Indian Ocean Island of Madagascar (Fig. 1). The vessel was carrying a cargo of rock phosphate fertiliser, along with bunker fuels and lubricating oil. Within a few days the vessel suffered serious structural deterioration as a result of its grounded position on the seabed, the strong sea-surge, and the breaking waves, so that its bulk cargo and its fuel and oil supplies could not be salvaged. Cap Sainte Marie is a remote location and is a special reserve famous for its coral reefs, rare species of tortoise and the migrating humpback whales that pass by at this time of year en route to their breeding grounds off Reunion Island. Madagascar is home to two percent of the world's biodiversity, and the majority of its plant and animal species are found nowhere else on earth. The fertiliser did not pose a severe threat to the local environment. The major threat came from the oils carried aboard the vessel; as a result of the incident 39,250 metric tonnes (MT) of cargo, 568 MT of heavy fuel oil, 66 MT of diesel oil, and 8 MT of lube oils were spilled into the sea. Most of the spillage was taken by currents to the east and away from the Cap Sainte Marie marine reserve and the coral reef, which minimised the environmental damage caused.

A large-scale clean-up operation was initiated and run by the owners of the vessel under expert guidance and using local workers. By March 2010, the clean-up works were completed to a high standard and operations were terminated with the approval of local, regional and national-level authorities. Following the incident, a number of scientific monitoring efforts were undertaken to investigate what impact the lost oil and cargo might have had on the local environment and population.

As part of an impact assessment, after the normal seasonal reopening of the lobster fishery, The International Tanker Owners Pollution Federation (ITOPF) undertook sampling of commercial species of fish and lobsters. These were obtained live from local fishermen operating in the coastal reef waters and tissue samples taken for PAH analysis. As far as possible, two samples of each taxon were collected at each station. The objective of the campaign was to collect samples along the entire coast of the Androy Region, both within and outside the affected area. A sample from the Fort Dauphin area was also collected as one of the control samples. The sampling and transport was undertaken with the aid of a helicopter, enabling the lobsters to be brought live and the fish fresh to Fort Dauphin. Slices of muscle tissue (i.e. the edible parts) were sampled from the fresh fish (posterior area to avoid sampling viscera) and from the live lobster (first segment of the tail). In all, tissues from 11 samples of fish (yellowfin bream – *Acanthropagrus australis*, known as Capitaine locally) and 42 samples of green and red lobsters (three species: *Panulirus homarus* ssp. *rubellus*, *P. penicillatus* and *P. Longipes*) were wrapped in aluminium foil, packed in small plastic containers sorted by location (with tags), frozen immediately and shipped to the UK by air freight. The samples were frozen immediately on arrival at the Centre for

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doi:10.1016/j.marpolbul.2011.09.019

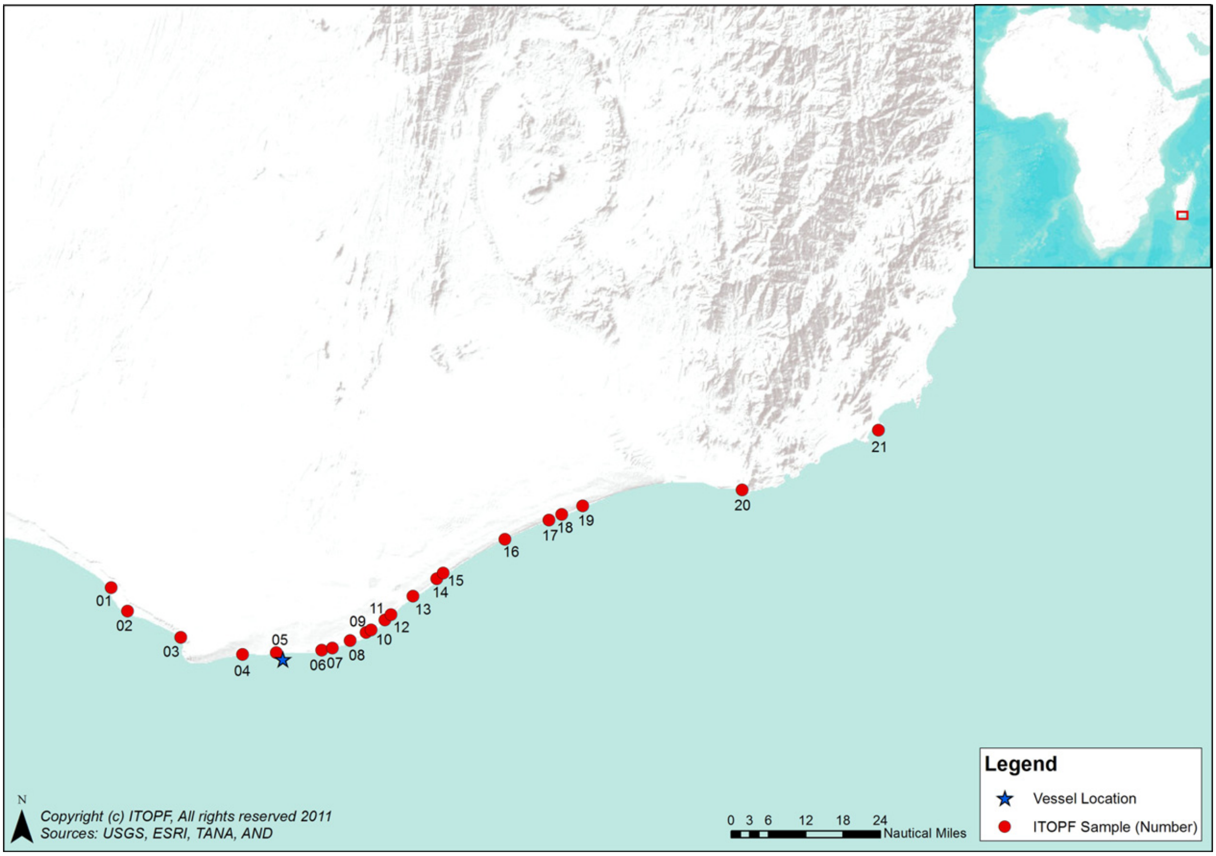

Fig. 1. Locations on the south coast of Madagascar from which samples were taken. The location of the wreck is indicated by a star.

Environment, Fisheries and Aquaculture Science (Cefas) and stored in a freezer at a temperature below -18 °C prior to analysis. Sampling locations are given in Table S1 in the electronic supplementary information available on the journal website.

Full details of the analytical methodology can be found in Kelly et al., 2000. Briefly, each homogenised tissue sample was extracted by alkaline saponification followed by liquid/liquid solvent extraction using glass-distilled grade pentane and drying of the extracts with sodium sulphate. The total hydrocarbon concentration in the extracts was determined by means of ultra-violet fluorescence spectrometry. Sample extracts were then reduced to 1 ml by means of a rotary-film evaporator, and then passed through a short alumina chromatography column in order to clean-up the extract, reconcentrated to 1 ml and sealed into an injection vial. A suite of parent and alkylated PAH were then determined using coupled gas chromatography–mass spectrometry (GC–MS) in full scan, electron impact (EI) ionisation mode. The GC–MS ion trap instrument was interfaced to and controlled by a data system using Xcalibur software. Quantification was by means of a range of deuterated internal standards added prior to sample extraction, with analytical quality control materials (certified or laboratory reference materials) being run within each sample batch. Results obtained for these reference materials were plotted on Shewhart control charts in order to monitor the day-to-day performance of the method, and were also used for acceptance/rejection of the batch data. In the event of the reference material data falling outside preset limits the batch would be rejected and reanalysed. In this study no batches were rejected. Participation in the QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) laboratory proficiency scheme also involves analysis of shellfish samples twice per year as an external quality check (Law et al., 2000). QUASIMEME require that, for data to be considered valid, the percentage of satisfactory result (z scores <2) should be >70%. This requirement was met.

A summary of the PAH data is given in Table 1, and the full dataset is presented in Tables S2 and S3 in the electronic supplementary information available on the journal website. $\sum$PAH concentrations (the sum of the concentrations of the PAH determined) ranged from 1.9 $\mu g\ kg^{-1}$ to 63 $\mu g\ kg^{-1}$ wet weight. No PAH of higher molecular weight than pyrene (i.e. greater than 202 Da) were detected (limit of quantification 0.1 $\mu g\ kg^{-1}$ wet weight per compound, or class in the case of alkylated compounds). Within the EU, limits have been set for benzo[*a*]pyrene in fish and shellfish (European Commission, 2006). Maximum permitted levels are 2.0 $\mu g\ kg^{-1}$ wet weight in fish muscle tissue and 5.0 $\mu g\ kg^{-1}$ wet weight in crustacean meat, excluding head and thorax meat of lobster. In addition, the UK has additional interim guideline limits for benz[*a*]anthracene and dibenz[*a,h*]anthracene of 15 $\mu g\ kg^{-1}$ wet weight per compound, to be applied following emergencies such as oil spills, where adequate background data do not exist to demonstrate that the concentrations found do not result from the emergency under consideration. The concentrations of these three PAH compounds were <0.1 $\mu g\ kg^{-1}$ wet weight in all samples. In addition, we calculated the summed PAH as benzo[*a*]pyrene equivalents (TEQ) (Law et al., 2002). In this case, rather than being summed directly, individual PAH concentrations are weighted in relation to their carcinogenic potential using toxic equivalency factors before being combined to yield an equivalent benzo[*a*]pyrene concentration reflecting the summed carcinogenicity of all compounds determined. In the current study, fluoranthene and pyrene were the only relevant compounds detected. Their concentrations were multiplied by factors of 0.02