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n-Paraffins in Polluted Fish by Crude Oil from "Juliana" Wreck

Terushige MOTOHIRO* and Norio INOUE**

Abstract

Samples of salmon, mullet and black sea bream were obtained from polluted water after the wreck of the ship "Juliana" in order to detect n-paraffins as a measure of crude oil contamination.

The results of gas chromatography of tissue from mullet and black sea bream revealed qualitative similarity consisting of 9 peaks all together, of which 8 were identical with those of C₁₃-C₂₀ n-paraffins. Little hydrocarbon was found in the muscle from salmon. The present results point to a possibility of determining crude oil contamination of fish from their paraffin content.

Introduction

In the stranding of the "Juliana" on the 30th November 1971, about 6,000 tons of crude oil were spilt on to the water outside Niigata Harbour, Niigata Pref. The oil polluted beach sand, rocks, sea weeds, and a large variety of fish and shellfish. After the spillage, complaints were received from fisherman that the fish in the set net were killed or that the fish they caught were inedible due to the presence of a crude oil taint. In order to try to investigate the contamination, samples of the fish were obtained from the polluted water for the detection of n-paraffins.¹⁾

Materials and Methods

Materials

One sample each of chum salmon, *Oncorhynchus keta*, mullet, *Mugil cephalus*, and black sea bream, *Mylio macrocephalus* was caught on the 5th December by set net which had been set on the 2nd December at the Tayuhama indicated in Fig. 1, and after frozen storage at the Niigata Prefectural Fisheries Experimental Station at -25°C for about 3 weeks, the samples were transferred to this laboratory in dry ice. A further sample each of *Mugil cephalus* and *Mylio macrocephalus* caught commercially off Hakodate were bought from fish market for comparative purposes.

Both oil contaminated and control *Mugil cephalus* and *Mylio macrocephalus*

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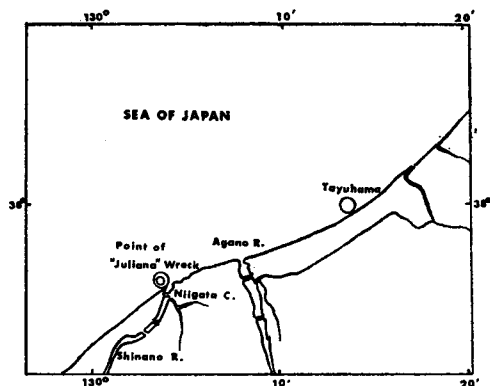


Fig. 1. Map of Niigata coastline showing point of "Juliana" wreck and Tayuhama.

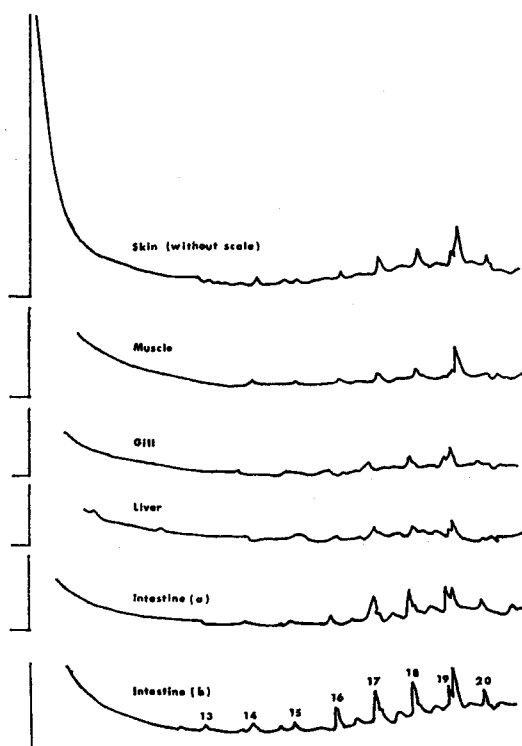


Fig. 2. Chromatograms of n-paraffins in the portions of black sea bream, *Mylio macrocephalus*. Numbers indicate the numbers of C-atoms in the chain of hydrocarbons.

were beheaded and deboned, and the tissues taken for investigation were skin without scale, muscle, liver, gill, and intestine. In addition, the pylorus was taken from *Mugil cephalus*. The intestine for the *Mylio macrocephalus* was cut into two portions (a) and (b) from which the contents were removed for investigation.

The tissue used in *Oncorhynchus keta* was muscle only.

Extraction and isolation of n-paraffins

To each portion was added 3 volumes of chloroform-methanol (2:1, v/v), and allowed to extract for 4 hours at room temperature after which it was filtered. The residue was extracted similarly for a further 16 hr. Both the extracts was washed with 0.05% NaCl and dehydrated with anhydrous sodium sulphate. The solvent was removed on a rotary evaporator under reduce pressure, the bath temperature being kept below 30°C.

The material obtained from the extraction was chromatographed on silicic acid (10 g) (100–200 mesh, Davison Chem. Co.) suspended in chloroform on a glass column (12 cm long, 10 mm id, holding volume 14 ml) and washed with 3 times volume of chloroform. The silicic acid had been previously activated by heating for 24 hr at 120°C. The extract was applied to the column in a minimum volume of chloroform and the n-paraffin fraction eluted with 45 ml of chloroform.

Thin layer chromatography was carried out on Wako Gel B-O with a solvent system consisting of n-hexane and benzene 1:1 by volume. The Wako Gel B-O used was activated by heating for 30 min at 105°C. The chromatographic fractionation was repeated three times in order to remove other lipid compounds. The hydrocarbon fraction was collected in a small column and eluted with chloroform, the solvent was removed and the product mixed with 160 μ l of chloroform, from which 3 μ l was applied to the gas chromatography.

Gas chromatography

The equipment used was a Shimadzu GC-4APF Gas Chromatograph fitted with a hydrogen flame ionization detector. The column was 2 m long and 3 mm id stainless steel in which 1.5% SE-30 (Chromosorb W, AW-DMCS, 60–80 mesh) was packed, operating at a flow rate of 35 ml/min for helium. The column temperature was programmed at 4°C/min from 70°C to 210°C.

Results and Discussion

The control and polluted samples supplied for the present experiment had a similar appearance and flavour, and did not possess any noticeable abnormal odour. However, the hydrocarbon fractions isolated from the polluted samples had strong crude oil odour, while the same fractions from the comparative samples were predominantly flavourless.

The resultant chromatograms (Figs. 2 and 3) revealed remarkable qualitative similarity. Of total 9 peaks, 8 were identical with standard pure C₁₃–C₂₀ n-paraffins, respectively. The peak found in both polluted and the comparative sample is not identified yet. It appears, however, that this peak might correspond

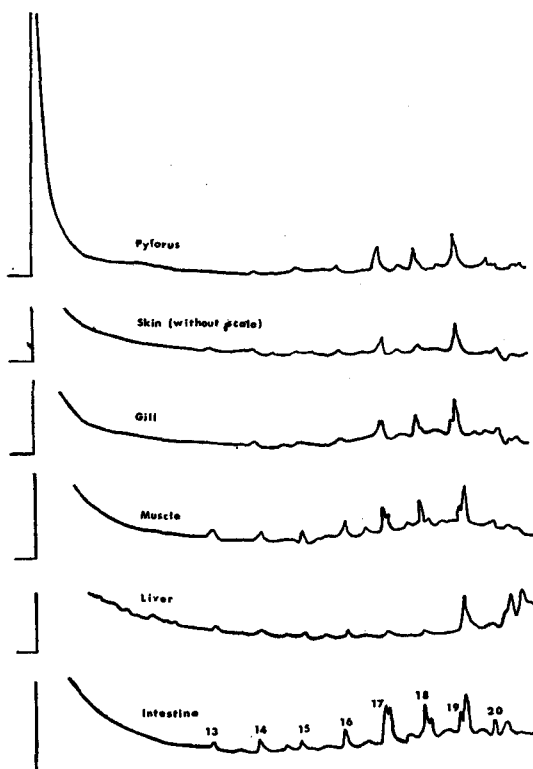


Fig. 3. Chromatograms of n-paraffins in the portions of mullet, *Mugil cephalus*. Numbers indicate the numbers of C-atoms in the chain of hydrocarbons.

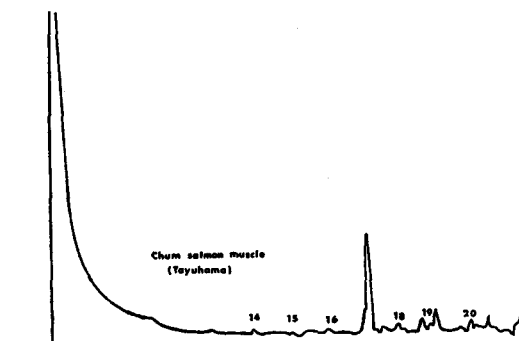


Fig. 4. Chromatogram of n-paraffins in the muscle of chum salmon, *Oncorhynchus keta*. Numbers indicate the numbers of C-atoms in the chain of hydrocarbons.

with natural hydrocarbon such as pristane.²⁾

The chromatograms shown in Figs. 2 and 3 suggest also that crude oil contamination could proceed from the surrounding water to organs of the fish through feeding,³⁾ because the muscle of the polluted sample of *Oncorhynchus keta*

contained few hydrocarbons (Fig. 4). This specimen was caught just before spawning and little material was found in the digestive tract.

The presence in and tainting of fish by petroleum hydrocarbons has been studied by a number of investigators,⁴⁾⁵⁾⁶⁾ and some kind of n-paraffins were identified in the hydrocarbon fraction isolated from the brown trout contaminated by diesel oil.²⁾ Unfortunately, there is little information on whether the detected hydrocarbons can produce a taint in fish, but only the present results and those of other workers²⁾ suggest the possibility of detecting crude oil contamination in fish by identifying n-paraffins.

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