

# Accumulation of Polycyclic Aromatic Hydrocarbons by Lobsters (*Homarus americanus*) Held in a Tidal Pound

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The levels of polycyclic aromatic hydrocarbons (PAH) in lobsters increased during 3-mo storage in a tidal pound constructed from creosoted lumber. For example, the concentrations of pyrene and benzo(a)pyrene in hepatopancreas of freshly landed lobsters were 59 and 6 ng/g (wet wt) respectively and increased to 4150 and 47 ng/g following 3-mo storage in winter and to 23 550 and 545 ng/g (wet wt) respectively following 3-mo storage in summer. The levels of these two PAH in tail muscle were 0.4 to 1.7% of the levels in hepatopancreas. Transfer of the lobsters to a creosote-free tidal pound resulted in decreases in pyrene and benzo(a)pyrene levels in hepatopancreas of 50 and 40% respectively over 5 wk in winter. Over 2 wk in summer, pyrene level decreased by 80% and change in benzo(a)pyrene ranged from -60 to +35%.

Key words: Lobster, creosote, polycyclic aromatic hydrocarbons, uptake, excretion, tidal pound

RÉSUMÉ

Uthe, J. F., D. W. McLeese, G. R. Sirota, and L. E. Burrige. 1984. Accumulation of polycyclic aromatic hydrocarbons by lobsters (Homarus americanus) held in a tidal pound. Can. Tech. Rep. Fish. Aquat. Sci. 1059: iii + 10 p.

Les concentrations d'hydrocarbures aromatiques polycycliques (HAP) dans des homards ont augmenté au cours d'un stockage d'une durée de trois mois dans un enceinte en bois créosoté. Par exemple, les concentrations de pyrène et de benzo(a)pyrène dans l'hépatopancréas d'homards fraîchement débarqués étaient de 59 ng/g et 6 ng/g (masse humide) respectivement et augmentaient à 4150 ng/g et 47 ng/g après un stockage d'une durée de trois mois en hiver, et à 23 550 ng/g et 545 ng/g (masse humide) respectivement après un stockage d'une durée de trois mois en été. Les concentrations de ces deux HAP dans le muscle de la queue correspondaient à 0,4 p. 100 et 1,7 p. 100 des concentrations dans l'hépatopancréas. Après transfert des homards dans une enceinte exempte de créosote, les concentrations de pyrène et de benzo(a) pyrène dans l'hépatopancréas ont diminué de 50 p. 100 et 40 p. 100 respectivement sur une période de cinq semaines en hiver. Sur une période de deux semaines en été, la concentration de pyrène a diminué de 80 p. 100, tandis que la concentration de benzo(a)pyrène a varié -60p. 100 à +35 p. 100.



## INTRODUCTION

Polycyclic aromatic hydrocarbons (PAH) are compounds with three or more fused aromatic rings. A complex mixture of PAH is present in fossil fuels and also is formed during combustion of carbonaceous fuels; the amount formed increases as the combustion efficiency decreases (National Academy of Sciences 1972). Concern over human exposure to PAH dates back to the eighteenth century when Pott (1775) associated an increased cancer incidence with chimney cleaning and soot handling. Volkman (1875) also found an association between the incidence of cancer and work in coal tar plants.

Coal tar, produced by the destructive distillation of bituminous coal, is redistilled to yield a number of fractions, including creosote, which is used in combination with naphthalene and coal tar to pressure-treat wood for preservation. In the marine environment, creosote is believed to be the best treatment available for preventing or delaying wood rot and wood-borer attack (Page 1977). Creosote must contain more than 98% aromatic components to be effective as a wood preservative in the marine environment (Baechler and Roth 1961). Both creosote and coal tar contain PAH at high levels (Colley 1974).

PAH contamination of marine shellfish living near marinas, harbors, or areas of oil spills has been known for many years (Cahnmann and Kuratsume 1957; Andelman and Suess 1970). Dunn and Stich (1975) found elevated levels of benzo(a)pyrene (BaP) in mussels (*Mytilus edulis*) taken from creosoted pilings. BaP is a carcinogenic PAH (National Academy of Sciences 1972). Zitko (1975) showed that mussels (*Mytilus edulis*), periwinkles (*Littorina littorea*) and various species of whelks had tissue PAH compositions closely resembling that of creosote. Sirota and Uthe (1981) showed the similarity of PAH patterns in creosote and lobster (*Homarus americanus*) digestive gland.

Dunn and Fee (1979) reported that lobsters held for approximately 3 mo in a commercial lobster storage tidal pound had high PAH levels in the hepatopancreas and tail muscle. PAH levels in hepatopancreas were 7-11 times higher than in tail muscle. They also found BaP in samples of other invertebrates such as clams, cockles, mussels, oysters and crabs, at levels between 1 and 10 ng/g. Levels of BaP greater than 1-10 ng/g are of concern to health officials since the usual level of BaP in most foodstuffs is less than 1 ng/g (Howard et al. 1968; Lo and Sandi 1978).

The purpose of this study was to follow changes in levels of PAH in lobsters held under typical summer and winter storage conditions in the same commercial tidal pound sampled previously by Dunn and Fee (1979). In part, the objective was to determine if conditions within the pound contributed to the accumulation of PAH within the lobsters. In addition, the ability of lobsters to eliminate accumulated PAH was assessed by transferring them to another holding facility where there was no creosote. Freshly caught lobsters from four sites in Nova Scotia were sampled to provide an estimate of background PAH levels in coastal stocks of lobsters.

## MATERIALS AND METHODS

## DETERMINATION OF PAH

Eleven non-alkylated PAH were measured: phenanthrene, fluoranthene, pyrene, triphenylene, benzo(a)anthracene, chrysene, benzo(e)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, and benzo(ghi)perylene. In the summer experiment, indeno(1,2,3-cd)pyrene (o-phenylene pyrene) levels also were determined.

PAH were determined by the method of Dunn and Armour (1980), with slight modifications. All solvents used were HPLC grade. Tissue homogenates were prepared and frozen in solvent-washed glass jars. Following KOH saponification, the residual lipids and PAH were extracted with iso-octane, then transferred to toluene for column chromatography on 30 g of 5% H<sub>2</sub>O deactivated Florisil. PAH were eluted with 200 mL of toluene, transferred to 5 mL of dimethyl sulfoxide (DMSO), and extracted into iso-octane from 66% aqueous DMSO, washed with water and then finally prepared for HPLC in 100  $\mu$ L DMSO. HPLC was carried out on a Vydac 201 TP reverse phase column (10  $\mu$ , 3.2 mm id, 25 cm L), using a linear gradient (70-94% acetonitrile/water [v/v] over 20 min). Instrumentation included two Waters M-6000 A pumps, a Waters WISP Model 710B sample processor, and Model 720 system controller. Column temperature was controlled at 25.0°C by an LC-22 column temperature controller (Bioanalytical Systems Inc.). Schoeffel variable wavelength detectors, Models SF770 (UV) and FS970 (fluorescence) were used in series and with separate chart recorders. Wavelengths used were as follows: UV = 265 and 254 nm; fluorescence ex = 280 nm; em = >389 nm. Both internal and external standardizations were used and peaks were identified by retention times, co-chromatography of reference compounds and, if necessary, by response ratios between UV and fluorescence detectors. In a few cases where the fluoranthene and pyrene peaks overlapped, pyrene was quantified by UV.

Recoveries of PAH added to tissue prior to saponification ranged from 83% for benzo(k)-fluoranthene to 109% for fluoranthene. Analytical variance (relative standard deviation) averaged 7.5%, ranging from 1.8% for benzo(e)pyrene to 13.6% for benzo(a)anthracene. N equaled 3 for each PAH.

## LOBSTER EXPOSURE

Ten lobsters were sampled from the same commercial tidal pound in Charlotte Co., New Brunswick, after approximately 1-, 2-, and 3-mo storage in winter (mid November-mid February) and after about 1-, 2-, 3, and 3.5-mo storage in summer (mid June-early September). At the times of the last samplings, 60 lobsters were transferred to a creosote-free holding facility in Shelburne Co., Nova Scotia. Samples of 10 lobsters were taken 1, 3, and 5 wk after transfer in winter, and at 18 h and at 4, 8, and 13 d after transfer in summer to follow excretion or depuration of the PAH. Length, weight, and sex of each lobster were recorded. The hepatopancreas and tail muscle were removed, weighed, and stored frozen in solvent-washed glass jars, each jar having an aluminum foil lid liner.

For analysis, equal weights of homogenized hepatopancreas from each of five animals were combined to yield two pooled samples from the 10 animals. Similarly, two samples of tail muscle were prepared. Animal-to-animal variation was determined by analyzing samples of hepatopancreas and tail muscle from each of 10 other lobsters taken on the last day of depuration in the summer experiment.

## RESULTS AND DISCUSSION

### BETWEEN ANIMAL VARIATION

For the 10 individual lobsters, the coefficients of variation (relative standard deviations) for the PAH ranged from 48-86% for hepatopancreas and from 21-70% for tail muscle (Table 1), and the coefficient for each PAH was greater for hepatopancreas than for tail muscle. The large variation among animals may be related to differences in the animals prior to capture, to exposure conditions within the tidal pound, and to metabolic differences among the animals.

The 10 animals ranged in weight from 429-1054 g, the mean weight being  $577 \pm 184$  g (mean  $\pm$  SD). Linear regression analysis did not reveal a significant ( $p = 0.05$ ) correlation between the concentration of PAH in tissue and the total weight of the animal.

The PAH levels in hepatopancreas averaged 35 times higher than those in tail muscle (Table 2). This is because the hepatopancreas, a digestive organ, has a higher fat level than tail muscle (Stewart et al. 1972) and may contain gut contents (van Weel 1979). Obviously, phenanthrene is distributed differently between tail and hepatopancreas since its mean ratio of 7.7 (Table 2) is lower than the mean ratios for the other 11 PAH which range from 29-52. Phenanthrene was the only tricyclic PAH measured and the low values for the ratio may be related to its relatively high water solubility (Neff 1979). Finally, it appears that there are substantial differences in the ratios between animals.

### UPTAKE AND DEPURATION OF PAH BY LOBSTERS

The concentrations of PAH in hepatopancreas and in tail muscle increased during storage, more PAH accumulating when the lobsters were at higher (summer) than at lower (winter) temperatures (Table 3, 4). For example, the summer to winter ratios of the PAH levels after 3-mo storage ranged from 2:1 to 19:1 for hepatopancreas and from 1:1 to 33:1 for tail muscle (Table 5).

PAH levels in tail muscle tended to increase for some time after the animals were moved to clean water (Table 3, 4), presumably because excretion was initiated and PAH was transported by blood from hepatopancreas to tail muscle and other tissues. However, holding the animals in clean water for longer periods resulted in lowering the PAH levels. Losses during 5 wk in winter ranged from 31-77% (mean 53%) in hepatopancreas and from 0-81% (mean 44%) in tail muscle. In summer, the results for excretion were variable. By 13 d, there were losses for only 4 of the 12 PAH in hepatopancreas (i.e. gains for 8 PAH) and losses for only 3 PAH in tail muscle. When transferred to clean water, lobsters had higher levels of PAH in summer than in winter,

and the excretion period for the summer lobsters was much shorter. From a total of 60 lobsters transferred, 50 had been sampled by 13 d. Mortality, in part caused by cannibalism, did not leave enough lobsters for a later sampling. Cannibalism during the summer excretion period was a factor that could alter the apparent excretion, assuming that PAH could be accumulated from food.

It was shown that PAH from creosote can accumulate rapidly in hepatopancreas of lobsters (McLeese and Metcalfe 1979). Lobsters were not sampled until 1 mo after they were placed in the tidal pound. Therefore, it is impossible to know whether, or how much, the conditions within the tidal pound contributed to the concentrations of PAH measured then. The mean PAH level after 1-mo storage was higher in winter than in summer in 17 of 22 comparisons. This could indicate that the winter stock of lobsters was more heavily contaminated initially. In support of this suggestion, there were decreases in the level of 7 of 11 PAH in hepatopancreas and in 8 of 11 PAH in tail muscle between the 1- and 2-mo periods in winter. In contrast, between 1- and 2-mo storage in summer there were increases in 23 of 24 PAH. Between 2- and 3-mo storage there were increases for each PAH for each tissue and season, indicating contribution of PAH from sediment or water in the tidal pound.

The ratios of the concentrations of a PAH to those of another PAH, for example pyrene/fluoranthene, can be used to show whether there are changes in the uptake or excretion of one PAH relative to the other. Also, the ratios can show whether uptake of PAH is proportional to its concentration in the environment.

Ratios of the concentration of some PAH (pyrene, benz(a)anthracene, and benzo(a)pyrene) to the concentration of fluoranthene were calculated for hepatopancreas and tail muscle data (Table 3, 4) and for sediment (Table 7). The mean ratios are summarized in Table 8.

Three outliers were eliminated from the calculation of the mean ratios for benzo(a)pyrene/fluoranthene for hepatopancreas in summer data (1- and 2-mo data) and from the corresponding data for tail muscle (Table 4). The outliers were more than three times the standard deviation for the mean ratio, with a maximum of two calculations of the mean ratio. Elimination of these values changed the mean ratio from 0.05 to 0.06 for hepatopancreas and from 0.06 to 0.07 for tail muscle.

There were no trends with time for any of the PAH ratios for hepatopancreas or tail muscle (Table 3, 4). Therefore, the concentrations of pyrene, benz(a)anthracene, and benzo(a)pyrene within the lobster tissues maintained the same proportional relationships with the concentration of fluoranthene throughout the uptake and excretion phases of the tests.

The mean ratios for pyrene/fluoranthene (4.1) and for benz(a)anthracene/fluoranthene (3.1) for hepatopancreas in winter are greater than the mean ratios for sediment, 1.1 and 1.3, respectively (Table 8). Presumably in winter, accumulation of these two PAH in hepatopancreas was enhanced relative to the accumulation of fluoranthene. With these exceptions, the general similarity of the ratios for PAH concentrations in lobster tissues and in sediment indicates uptake of these four PAH was

proportional to their concentrations in the sediment.

The levels of PAH were low for lobsters captured at four sites around Nova Scotia (Table 6) and, as expected, the levels in tail muscle often were immeasurable and lower than in hepatopancreas. Compared with data for New Harbour lobsters, the PAH levels in lobsters after 3-mo storage in the tidal pound were 2-60 times higher in winter (Table 3) and 13-170 times higher in summer (Table 4).

Dunn and Fee (1979) reported the range of tail muscle levels of the carcinogen BaP to be 0.4-0.9 ng/g wet wt for lobsters prior to storage. After storage in the tidal pound for approximately 3 mo, BaP levels in tail muscle ranged from 7.4-281 ng/g. In our study, levels of BaP in tail muscle ranged from trace amounts to 1.5 ng/g in control lobsters (Table 6). The mean value after storage in the tidal pound was 0.2 ng/g after 3 mo in winter and was 6.5 and 7.5 ng/g after 3 and 3½ mo respectively in summer. None of these values approached the maximum value of 281 ng/g found by Dunn and Fee (1979). In hepatopancreas the BaP levels ranged from 0.8-22 ng/g for control lobsters (Table 6) and the mean value was 46.5 ng/g after 3 mo in winter and was 507 and 361 ng/g after 3 and 3½ mo respectively in summer (Table 3, 4).

Sediments from the tidal pound were analyzed for PAH (Table 7). Values for phenanthrene, fluoranthene, and pyrene in the tidal pound sediments, ranged from 650-3100 ng/g (Table 7) and were higher than those in a 215-m deep sediment from the Gulf of Maine (43-120 ng/g, wet weight) (Laflamme and Hites 1978), or for those in a sediment 45 km seaward from Boston, Mass., at 90 m deep (250-360 ng/g). However, the tidal pound sediment values were similar to or lower than values for Boston harbor or Charles River sediments which ranged from 670-13,000 ng/g (Windsor and Hites 1979). Levels similar to or slightly higher than those in the tidal pound sediments were reported for several PAH by John et al. (1979) for sediments from the Severn Estuary, Great Britain, an area characterized by high tides and receiving industrial effluents. The elevated PAH levels in the sediment of the tidal pound probably resulted from the creosoted lumber used in the construction of the pound.

The uptake of PAH by impounded lobster may be enhanced because lobsters tend to burrow into sediments (McLeese and Wilder 1964), eat each other, and a few may gnaw at the wooden parts of the enclosures. Because of the possibility of tainting, pound owners have been warned against storage of lobster in freshly creosoted pounds (McLeese and Wilder 1964). However, there is no evidence that lobsters are tainted after being held in creosoted structures which have been exposed to sea water for extended periods. Most of the creosote loss from creosoted wood to sea water occurs during the first year (Gibb 1978). Creosoted timbers in current tidal pounds are likely to be several years old, particularly since it is reported that recent repairs are made with timbers pressure-treated with a non-creosote preservative. The relative roles of old creosote timbers and of contaminated sediment as they affect uptake of PAH by lobsters in storage are not fully understood. Assuming contaminated sediment may be a continuing major source of PAH,

the extent and distribution (area and depth) of the PAH-contaminated sediments in and around each tidal pound will require assessment before sediment removal can be recommended.

There are 18 tidal pounds in New Brunswick (Bay of Fundy area) and three in southwestern Nova Scotia (total 21). According to a recent survey conducted by questionnaires, four tidal pounds had minor amounts of creosoted materials and 17 were reported to be constructed with untreated wood (no creosote) (McLeese 1983). An earlier survey indicated that six pounds had minor amounts of creosoted materials, two had been constructed many years ago with creosoted material and that 13 were constructed with untreated material (McLeese 1983). The potential for lobsters in tidal pounds to become contaminated with PAH probably is limited to a few of the 21 tidal pounds.

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Table 1. PAH concentrations (ng/g wet weight) in hepatopancreas and tail muscle of 10 individual lobsters following 3 mo summer holding in a creosoted pound, then transferred to clean water for 13 d.

| PAH                    | Lobster no. |      |      |      |      |       |       |       |       |       | C V <sup>a</sup> | Mean  |
|------------------------|-------------|------|------|------|------|-------|-------|-------|-------|-------|------------------|-------|
|                        | 1           | 2    | 3    | 4    | 5    | 6     | 7     | 8     | 9     | 10    |                  |       |
| <u>Hepatopancreas</u>  |             |      |      |      |      |       |       |       |       |       |                  |       |
| Phenanthrene           | 1980        | 2000 | 720  | 700  | 1100 | 3400  | 390   | 1600  | 3100  | 3000  | 66               | 1799  |
| Fluoranthene           | 3800        | 6700 | 3400 | 2100 | 3400 | 10300 | 6500  | 7100  | 10600 | 8500  | 48               | 6240  |
| Pyrene                 | 1300        | 3000 | 1200 | 420  | 1400 | 5500  | 1800  | 2400  | 2700  | 3500  | 63               | 2322  |
| Triphenylene           | 11500       | 8200 | 7800 | 4100 | 6700 | 32800 | 15200 | 24700 | 39000 | 18700 | 69               | 16870 |
| Benz(a)anthracene      | 6200        | 8100 | 4900 | 2000 | 4500 | 20000 | 9500  | 17500 | 19400 | 1960  | 64               | 9406  |
| Chrysene               | 5600        | 5300 | 3600 | 1800 | 3100 | 17800 | 7200  | 16100 | 16700 | 17600 | 70               | 9480  |
| Benzo(e)pyrene         | 1200        | 500  | 360  | 210  | 250  | 1600  | 780   | 1200  | 1400  | 1200  | 59               | 870   |
| Benzo(b)fluoranthene   | 790         | 620  | 440  | 370  | 390  | 2000  | 950   | 2100  | 1800  | 1800  | 63               | 1127  |
| Benzo(k)fluoranthene   | 200         | 160  | 110  | 100  | 100  | 530   | 250   | 520   | 450   | 440   | 63               | 286   |
| Benzo(a)pyrene         | 220         | 150  | 140  | 100  | 140  | 860   | 270   | 830   | 680   | 740   | 78               | 413   |
| Benzo(ghi)perylene     | 340         | 240  | 240  | 140  | 240  | 1600  | 400   | 1700  | 1300  | 1400  | 86               | 760   |
| Indeno(1,2,3-cd)pyrene | 1000        | 690  | 560  | 390  | 500  | 4300  | 1300  | 4300  | 2900  | 3300  | 83               | 1924  |
| <u>Tail muscle</u>     |             |      |      |      |      |       |       |       |       |       |                  |       |
| Phenanthrene           | 300         | 140  | 280  | 270  | 180  | 210   | 220   | 260   | 310   | 200   | 21               | 237   |
| Fluoranthene           | 91          | 97   | 180  | 99   | 67   | 180   | 200   | 210   | 230   | 240   | 40               | 159   |
| Pyrene                 | 10          | 35   | 56   | 22   | 29   | 83    | 47    | 57    | 61    | 78    | 47               | 48    |
| Triphenylene           | 280         | 150  | 430  | 320  | 200  | 650   | 550   | 820   | 320   | 590   | 48               | 431   |
| Benz(a)anthracene      | 200         | 210  | 260  | 130  | 100  | 370   | 410   | 560   | 310   | 370   | 56               | 292   |
| Chrysene               | 190         | 130  | 160  | 110  | 60   | 290   | 280   | 430   | 200   | 250   | 50               | 210   |
| Benzo(e)pyrene         | 30          | 14   | 20   | 10   | 10   | 36    | 30    | 45    | 22    | 30    | 45               | 25    |
| Benzo(b)fluoranthene   | 30          | 20   | 26   | 27   | 9    | 42    | 46    | 72    | 31    | 35    | 48               | 34    |
| Benzo(k)fluoranthene   | 7           | 4    | 7    | 7    | 2    | 11    | 13    | 19    | 9     | 10    | 51               | 9     |
| Benzo(a)pyrene         | 8           | 5    | 10   | 12   | 4    | 21    | 19    | 32    | 15    | 16    | 57               | 14    |
| Benzo(ghi)perylene     | 15          | 8    | 15   | 17   | 6    | 39    | 29    | 59    | 24    | 25    | 64               | 24    |
| Indeno(1,2,3-cd)pyrene | 45          | 21   | 34   | 44   | 12   | 100   | 78    | 170   | 60    | 59    | 70               | 62    |

<sup>a</sup>Coefficient of variation.

Table 2. Ratio of PAH concentrations of hepatopancreas to tail muscle in individual lobsters following 3 mo summer holding in creosoted pound, then transferred to clean water for 13 d.

| PAH <sup>a</sup>                 | Lobster no. |       |      |      |       |       |      |      |       |       | Mean ratio ± SD |
|----------------------------------|-------------|-------|------|------|-------|-------|------|------|-------|-------|-----------------|
|                                  | 1           | 2     | 3    | 4    | 5     | 6     | 7    | 8    | 9     | 10    |                 |
| Phenanthrene                     | 6.6         | 14    | 2.6  | 2.6  | 6.1   | 16    | 1.8  | 6.3  | 9.9   | 15    | 7.7 ± 6.6       |
| Fluoranthene                     | 42          | 69    | 20   | 21   | 50    | 58    | 32   | 33   | 45    | 36    | 40 ± 16         |
| Pyrene                           | 130         | 86    | 21   | 19   | 48    | 67    | 37   | 42   | 44    | 45    | 52 ± 29         |
| Triphenylene                     | 42          | 54    | 18   | 13   | 34    | 50    | 28   | 30   | 123   | 32    | 42 ± 31         |
| <u>Benzo(a)anthracene</u>        | 31          | 38    | 19   | 15   | 45    | 54    | 23   | 31   | 62    | 5.3   | 37 ± 16         |
| <u>Chrysene</u>                  | 29          | 41    | 23   | 16   | 52    | 61    | 25   | 37   | 82    | 70    | 44 ± 22         |
| <u>Benzo(e)pyrene</u>            | 40          | 36    | 18   | 21   | 25    | 44    | 26   | 28   | 65    | 41    | 35 ± 14         |
| <u>Benzo(b)fluoranthene</u>      | 26          | 31    | 17   | 14   | 43    | 49    | 20   | 29   | 57    | 52    | 34 ± 15         |
| <u>Benzo(k)fluoranthene</u>      | 28          | 40    | 17   | 15   | 50    | 48    | 20   | 27   | 50    | 46    | 33 ± 14         |
| <u>Benzo(a)pyrene</u>            | 27          | 30    | 14   | 8    | 35    | 42    | 14   | 26   | 37    | 46    | 29 ± 14         |
| <u>Benzo(ghi)perylene</u>        | 23          | 30    | 16   | 8    | 39    | 41    | 14   | 29   | 53    | 57    | 31 ± 16         |
| <u>Indeno (1,2,3-cd)pyrene</u>   | 22          | 33    | 16   | 9    | 43    | 43    | 16   | 25   | 48    | 56    | 31 ± 16         |
| All PAH mean ratio ± SD          | 36±26       | 42±19 | 17±6 | 14±6 | 39±13 | 48±13 | 21±9 | 29±9 | 57±27 | 45±14 | 35              |
| Carcinogenic PAH mean ratio ± SD | 30±7        | 35±4  | 18±3 | 14±5 | 41±9  | 49±7  | 21±5 | 30±5 | 60±13 | 53±10 |                 |

<sup>a</sup>Carcinogenic PAH are underlined.

Table 3. PAH concentrations (ng/g wet weight) in hepatopancreas and tail muscle of lobsters held in a tidal pound in winter, then transferred to a creosote-free storage facility. The two entries under each date are values from analysis of two pooled tissue samples from five animals each.

| PAH                                  | Uptake         |      |                |       |                |       | Excretion      |       |               |       |                |      |
|--------------------------------------|----------------|------|----------------|-------|----------------|-------|----------------|-------|---------------|-------|----------------|------|
|                                      | Dec. 13 (1 mo) |      | Jan. 14 (2 mo) |       | Feb. 13 (3 mo) |       | Feb. 21 (1 wk) |       | Mar. 6 (3 wk) |       | Mar. 21 (5 wk) |      |
| <u>Hepatopancreas</u>                |                |      |                |       |                |       |                |       |               |       |                |      |
| Phenanthrene                         | 1130           | 1120 | 1580           | 1122  | 2500           | 2450  | 2300           | 1340  | 1600          | 1630  | 1320           | 2090 |
| Fluoranthene                         | 450            | 295  | 601            | 583   | 689            | 1050  | 587            | 633   | 677           | 670   | 464            | 370  |
| Pyrene                               | 1760           | 1380 | 2050           | 1850  | 2910           | 5400  | 2390           | 2320  | 2520          | 2450  | 2330           | 1850 |
| Triphenylene                         | 3560           | 3650 | 3040           | 3320  | 6570           | 11300 | 5760           | 4930  | 5630          | 4800  | 5340           | 4790 |
| Benz(a)anthracene                    | 1360           | 1070 | 1590           | 1780  | 2430           | 3650  | 2210           | 2090  | 1910          | 1730  | 1360           | 1280 |
| Chrysene                             | 349            | 400  | 170            | 250   | 626            | 748   | 534            | 426   | 333           | 267   | 344            | 412  |
| Benzo(e)pyrene                       | 528            | 723  | 302            | 538   | 1150           | 2000  | 1150           | 785   | 608           | 489   | 497            | 655  |
| Benzo(b)fluoranthene                 | 108            | 119  | 70             | 98    | 223            | 342   | 213            | 139   | 127           | 92    | 94             | 115  |
| Benzo(k)fluoranthene                 | 22             | 22   | 14             | 21    | 49             | 66    | 47             | 27    | 23            | 18    | 17             | 22   |
| Benzo(a)pyrene                       | 33             | 46   | 9              | 17    | 56             | 37    | 63             | 32    | 23            | 16    | 20             | 36   |
| Benzo(ghi)perylene                   | 117            | 141  | 23             | 58    | 283            | 576   | 333            | 179   | 129           | 100   | 74             | 127  |
| Indeno (1,2,3-cd)pyrene <sup>a</sup> | -              | -    | -              | -     | -              | -     | -              | -     | -             | -     | -              | -    |
| <u>Ratios</u>                        |                |      |                |       |                |       |                |       |               |       |                |      |
| Pyrene/fluoranthene                  | 3.8            | 4.8  | 3.4            | 3.2   | 4.2            | 5.3   | 4.0            | 3.7   | 3.7           | 3.7   | 5.0            | 5.0  |
| Benz(a)anthracene/fluoranthene       | 3.0            | 3.6  | 2.6            | 3.0   | 3.6            | 3.4   | 3.7            | 3.3   | 2.9           | 2.6   | 2.9            | 3.3  |
| Benzo(a)pyrene/fluoranthene          | 0.07           | 0.16 | 0.01           | 0.03  | 0.08           | 0.04  | 0.1            | 0.05  | 0.03          | 0.02  | 0.04           | 0.1  |
| <u>Tail muscle</u>                   |                |      |                |       |                |       |                |       |               |       |                |      |
| Phenanthrene                         | 20             | 36   | 55             | 35    | 59             | 95    | 108            | 110   | 93            | 77    | 42             | 6    |
| Fluoranthene                         | 45             | 41   | 15             | 19    | 36             | 48    | 61             | 86    | 31            | 23    | 17             | 22   |
| Pyrene                               | 45             | 42   | 54             | 42    | 53             | 87    | 98             | 123   | 66            | 36    | 19             | 8    |
| Triphenylene                         | 86             | 137  | 89             | 165   | 109            | 172   | 278            | 261   | 134           | 70    | 61             | 135  |
| Benz(a)anthracene                    | 62             | 100  | 49             | 47    | 96             | 133   | 201            | 226   | 80            | 56    | 48             | 60   |
| Chrysene                             | 8              | 14   | 5              | 5     | 10             | 16    | 29             | 21    | 10            | 5     | 4              | 9    |
| Benzo(e)pyrene                       | 18             | 39   | 13             | 13    | 21             | 35    | 99             | 53    | 29            | 13    | 11             | 26   |
| Benzo(b)fluoranthene                 | 5              | 9    | 4              | 4     | 7              | 11    | 21             | 16    | 9             | 4     | 4              | 6    |
| Benzo(k)fluoranthene                 | 0.9            | 1.6  | 0.7            | 0.6   | 1.2            | 2.1   | 4              | 3     | 1.5           | 0.6   | 0.6            | 1    |
| Benzo(a)pyrene                       | 0.5            | 2.8  | 0.1            | 0.1   | 0.2            | 0.2   | 1.8            | 0.4   | 0.6           | 0.1   | 0.1            | 0.3  |
| Benzo(ghi)perylene                   | 2              | 8    | 1.6            | 1.7   | 5              | 6     | 37             | 13    | 9             | 4     | 2.5            | 7    |
| Indeno (1,2,3-cd)pyrene <sup>a</sup> | -              | -    | -              | -     | -              | -     | -              | -     | -             | -     | -              | -    |
| <u>Ratios</u>                        |                |      |                |       |                |       |                |       |               |       |                |      |
| Pyrene/fluoranthene                  | 1.0            | 1.0  | 3.7            | 2.7   | 1.5            | 1.8   | 1.6            | 1.4   | 2.1           | 1.5   | 1.1            | 0.4  |
| Benz(a)anthracene/fluoranthene       | 1.4            | 2.4  | 3.2            | 2.5   | 2.6            | 2.8   | 3.3            | 2.6   | 2.6           | 2.4   | 2.8            | 2.7  |
| Benzo(a)pyrene/fluoranthene          | 0.01           | 0.07 | 0.006          | 0.005 | 0.005          | 0.004 | 0.03           | 0.004 | 0.2           | 0.004 | 0.006          | 0.01 |

<sup>a</sup>Not measured in winter experiment.

Table 4. PAH concentrations (ng/g wet weight) in hepatopancreas and tail muscle of lobsters held in a tidal pond in summer, then transferred to a creosote-free storage facility. The two entries under each date are values from analysis of two pooled tissue samples from five animals each.

| PAH                            | Uptake            |                   |                   |                     |                   | Excretion        |                   |                    |       |       |       |       |       |      |       |
|--------------------------------|-------------------|-------------------|-------------------|---------------------|-------------------|------------------|-------------------|--------------------|-------|-------|-------|-------|-------|------|-------|
|                                | June 17<br>(1 mo) | July 24<br>(2 mo) | Aug. 19<br>(3 mo) | Sept. 4<br>(3.5 mo) | Sept. 5<br>(18 h) | Sept. 8<br>(4 d) | Sept. 13<br>(8 d) | Sept. 17<br>(13 d) |       |       |       |       |       |      |       |
| Phenanthrene                   | 1800              | 1280              | 3350              | 7250                | 9010              | 3990             | 3670              | 4410               | 4360  | 7110  | 6780  | 1620  | 3390  | 1270 | 2460  |
| Fluoranthene                   | 406               | 326               | 4930              | 2700                | 9410              | 7810             | 2850              | 7810               | 11500 | 8610  | 9640  | 3380  | 3780  | 3610 | 8400  |
| Pyrene                         | 2200              | 1150              | 3550              | 1640                | 15900             | 31200            | 20800             | 10700              | 13200 | 16600 | 4040  | 5420  | 6610  | 2980 | 3520  |
| Triphenylene                   | 1370              | 763               | 5050              | 3250                | 11500             | 33200            | 11700             | 10700              | 25000 | 28700 | 16400 | 13500 | 16800 | 9900 | 29100 |
| Benz(a)anthracene              | 323               | 303               | 5630              | 2570                | 14800             | 18300            | 13000             | 12700              | 14000 | 15600 | 9200  | 7232  | 7890  | 5510 | 17400 |
| Chrysene                       | 576               | 347               | 4060              | 1670                | 11700             | 14900            | 10600             | 8350               | 11200 | 13200 | 7100  | 6480  | 7920  | 3950 | 15500 |
| Benzo(e)pyrene                 | 178               | 120               | 1880              | 369                 | 5160              | 3570             | 649               | 660                | 998   | 1190  | 1130  | 602   | 600   | 372  | 1230  |
| Benzo(b)fluoranthene           | 75                | 37                | 503               | 191                 | 1240              | 1690             | 1140              | 1020               | 1240  | 1590  | 1340  | 841   | 758   | 564  | 1760  |
| Benzo(k)fluoranthene           | 17                | 8                 | 140               | 51                  | 341               | 444              | 313               | 258                | 337   | 431   | 348   | 176   | 186   | 145  | 447   |
| Benzo(a)pyrene                 | 0.9               | 1.8               | 180               | 29                  | 407               | 680              | 420               | 303                | 488   | 651   | 332   | 308   | 336   | 158  | 710   |
| Benzo(ghi)perylene             | 29                | 18                | 366               | 86                  | 898               | 1180             | 624               | 490                | 827   | 1120  | 942   | 474   | 636   | 258  | 1370  |
| Indeno(1,2,3-cd)pyrene         | 67                | 39                | 607               | 156                 | 1540              | 2080             | 1090              | 769                | 1360  | 1860  | 2260  | 1110  | 788   | 743  | 3200  |
| Pyrene/fluoranthene            | 0.515             | 3.6               | 0.7               | 0.6                 | 1.7               | 2.7              | 1.3               | 2.8                | 1.4   | 1.1   | 1.9   | 0.4   | 1.6   | 1.7  | 0.8   |
| Benz(a)anthracene/fluoranthene | 0.8               | 0.9               | 1.1               | 1.5                 | 1.7               | 1.6              | 1.5               | 1.7                | 1.7   | 1.5   | 1.8   | 0.7   | 1.7   | 1.5  | 0.4   |
| Benzo(a)pyrene/fluoranthene    | 0.002             | 0.005             | 0.04              | 0.01                | 0.04              | 0.06             | 0.05              | 0.04               | 0.06  | 0.06  | 0.07  | 0.03  | 0.09  | 0.04 | 0.08  |
| Phenanthrene                   | 117               | 20                | 62                | 71                  | 82                | 104              | 110               | 19                 | 129   | 117   | 262   | 100   | 55    | 260  | 184   |
| Fluoranthene                   | 20                | 31                | 28                | 44                  | 68                | 111              | 160               | 69                 | 147   | 117   | 97    | 110   | 189   | 213  | 94    |
| Pyrene                         | 59                | 11                | 81                | 74                  | 154               | 221              | 35                | 510                | 206   | 206   | 94    | 191   | 194   | 42   | 83    |
| Triphenylene                   | 33                | 11                | 64                | 51                  | 182               | 239              | 302               | 198                | 311   | 253   | 315   | 423   | 603   | 562  | 311   |
| Benz(a)anthracene              | 43                | 10                | 71                | 51                  | 185               | 291              | 334               | 206                | 311   | 323   | 209   | 237   | 389   | 350  | 177   |
| Chrysene                       | 32                | 12                | 67                | 42                  | 156               | 226              | 258               | 221                | 204   | 230   | 132   | 210   | 288   | 253  | 116   |
| Benzo(e)pyrene                 | 6.8               | 3                 | 16                | 8.8                 | 22                | 22               | 37                | 26                 | 24    | 43    | 26    | 27    | 13    | 30   | 19    |
| Benzo(b)fluoranthene           | 4                 | 2                 | 8                 | 7                   | 19                | 30               | 35                | 27                 | 27    | 35    | 18    | 26    | 41    | 44   | 22    |
| Benzo(k)fluoranthene           | 0.9               | 0.5               | 1.5               | 1.8                 | 4                 | 6.4              | 9                 | 4                  | 7.5   | 8     | 4.7   | 5.8   | 11    | 11   | 5     |
| Benzo(a)pyrene                 | 0.1               | 0.1               | 0.4               | 0.2                 | 5                 | 8                | 10                | 5                  | 7     | 6.7   | 5.7   | 9.5   | 20    | 19   | 7     |
| Benzo(ghi)perylene             | 1.6               | 0.8               | 4.6               | 2                   | 14                | 21               | 20                | 12                 | 15    | 19    | 86    | 17    | 33    | 28   | 11    |
| Indeno(1,2,3-cd)pyrene         | 3.6               | 2.5               | 17                | 8                   | 43                | 60               | 37                | 59                 | 66    | 66    | 28    | 47    | 80    | 72   | 29    |
| Pyrene/fluoranthene            | 1.9               | 0.5               | 2.9               | 1.7                 | 2.3               | 1.5              | 1.4               | 0.5                | 3.7   | 1.4   | 1.0   | 1.7   | 1.0   | 0.9  | 0.4   |
| Benz(a)anthracene/fluoranthene | 1.4               | 2.0               | 2.6               | 1.2                 | 2.7               | 2.5              | 2.1               | 3.0                | 2.3   | 2.2   | 2.2   | 2.2   | 2.0   | 1.6  | 2.1   |
| Fluoranthene                   | 0.003             | 0.005             | 0.01              | 0.004               | 0.07              | 0.07             | 0.06              | 0.07               | 0.05  | 0.04  | 0.06  | 0.08  | 0.1   | 0.09 | 0.07  |
| Benzo(a)pyrene/fluoranthene    | 0.004             | 0.005             | 0.01              | 0.004               | 0.07              | 0.07             | 0.06              | 0.07               | 0.05  | 0.04  | 0.06  | 0.08  | 0.1   | 0.09 | 0.07  |

<sup>a</sup>Trace. Underlined values, outliers eliminated from calculation of mean.

Table 5. Ratio of summer/winter concentrations of PAH in each of two tissues of lobsters after storage in a tidal pound for 3 mo.

| PAH <sup>a</sup>     | Ratios for hepatopancreas | Ratios for tail muscle |
|----------------------|---------------------------|------------------------|
| Phenanthrene         | 3                         | 1                      |
| Fluoranthene         | 12                        | 2                      |
| Pyrene               | 6                         | 2                      |
| Triphenylene         | 3                         | 2                      |
| Benz(a)anthracene    | 5                         | 2                      |
| Chrysene             | 19                        | 15                     |
| Benzo(e)pyrene       | 3                         | 1                      |
| Benzo(b)fluoranthene | 5                         | 2                      |
| Benzo(k)fluoranthene | 7                         | 3                      |
| Benzo(a)pyrene       | 12                        | 33                     |
| Benzo(ghi)perylene   | 2                         | 3                      |

<sup>a</sup>Concentrations in Tables 3 and 4.

Table 6. Mean PAH concentrations (ng/g wet weight) in hepatopancreas and tail muscle of freshly captured lobsters from four locations around Nova Scotia; number of lobsters sampled indicated in brackets.

| PAH                     | Port Hood (14)  |             | Cape Sable (10) |                 | New Harbour (10) |             | East River (10) |                |
|-------------------------|-----------------|-------------|-----------------|-----------------|------------------|-------------|-----------------|----------------|
|                         | Hepato-pancreas | Tail muscle | Hepato-pancreas | Tail muscle     | Hepato-pancreas  | Tail muscle | Hepato-pancreas | Tail muscle    |
| Phenanthrene            | 100             | 7           | 14              | nd <sup>a</sup> | 94               | 16          | 27              | T <sup>b</sup> |
| Fluoranthene            | 150             | 5           | 19              | 4               | 400              | 28          | 41              | 7.5            |
| Pyrene                  | T               | nd          | 95              | nd              | 140              | 5           | T               | nd             |
| Triphenylene            | 55              | 2           | nd              | nd              | 150              | 7           | nd              | nd             |
| Benz(a)anthracene       | 22              | T           | 43              | 16              | 600              | 27          | 8.5             | T              |
| Chrysene                | 11              | nd          | 36              | 12              | 240              | 2           | 4               | T              |
| Benzo(e)pyrene          | 20              | T           | 15              | 4               | 340              | 4           | 12              | T              |
| Benzo(b)fluoranthene    | 6.5             | 0.4         | 47              | 2               | 45               | 3.5         | 16              | T              |
| Benzo(k)fluoranthene    | 1.5             | T           | 7               | 0.2             | 13               | 0.9         | 1.0             | T              |
| Benzo(a)pyrene          | 0.8             | T           | 1               | T               | 22               | 1.5         | 0.8             | T              |
| Benzo(ghi)perylene      | 6.7             | 0.5         | 21              | 0.3             | 16               | 1.5         | 2.5             | T              |
| Indeno (1,2,3-cd)pyrene | 4.2             | 0.2         | 47              | 3.0             | 45               | 1.5         | 5.0             | 0.3            |

<sup>a</sup>Not detected.

<sup>b</sup>Trace.

Port Hood, Inverness Co.; Cape Sable, Shelburne Co.; New Harbour, Guysborough Co.; East River, Lunenburg Co.; lobsters captured in May.