

Fate of Petroleum Hydrocarbons in Sewage Sludge after Land Disposal

D. Liu

National Water Research Institute, Burlington, Ontario, Canada L7R 4A6

With many sewage treatment plants achieving better removal of pollutants, the problem of disposing of the resulting sewage sludge is becoming one of the most important considerations to be faced in the management of total environment. In fact, sludge processing and disposal account for the largest single share on the total operation cost of the municipal wastewater treatment plant (MICHEL 1970). Almost every major city in the world is now facing the difficult problem of sludge disposal.

Sludge is normally disposed of by means of ocean dumping, incineration, or land disposal. However, the practice of ocean dumping is not recommended because of the potential for rapid accumulation of heavy metals and toxic organic substances by aquatic organisms (EMBER 1975). Incineration is also no longer considered to be desirable because of the energy cost and the air pollution problems that are often associated with it as well as the possibility of volatilization of persistent contaminants. Therefore, sewage sludge recycling by farmland disposal is becoming more attractive as a means to solve this disposal problem.

In the application of sewage sludge to farmland, a crucial factor to be considered is the possibility of accumulating heavy metals and other toxic organic substances in the agricultural soil systems. While information regarding the cumulative effect of heavy metals in sewage sludge on farm soils is available (LINNMAN et al. 1973, CUNNINGHAM et al. 1975, PIKE et al. 1975), little information is available concerning the fate of organic substances in sewage sludge, particularly after land disposal on agricultural soil systems.

Petroleum hydrocarbons are fairly resistant to the biological treatment process in sewage plants; consequently, high concentrations of petroleum hydrocarbons have been detected in various sewage sludges (LOEHR and HIGGINS 1965, LOEHR and DE NARARRA 1969, SHAW 1975). Petroleum hydrocarbons are known to be toxic to plants and crops because of their damaging effect on the processes of translocation, transpiration and photosynthesis (BAKER 1970). The present study was intended to investigate the fate of petroleum hydrocarbons in digested sewage sludge after farmland disposal.

Information so obtained would be valuable for setting up guidelines for sludge land disposal.

MATERIALS AND METHODS

During a 1972-73 characterization study of municipal sewage sludges in Ontario, it was noticed that the sludge from the Point Edward Water Pollution Control Plant consistently had a very high level of lipids and petroleum hydrocarbons (LIU 1978). Experiments, therefore, were set up to examine the fate of petroleum hydrocarbons in this particular sludge after land disposal using a lysimeter system. The fiberglass lysimeter had a depth of 183 cm and a diameter of 30.5 cm. The lysimeters were installed in an environmentally-controlled trailer with their tops projecting 7.6 cm above the trailer's roof to ensure their exposure to normal ambient and atmospheric conditions. Silt loam from Elora (Ontario) was used to build the lysimeter due to its being the predominant agricultural soil in southwestern Ontario. The soil had a pH of 7.8 with 3.6% organic matter. Its bulk density was approximately 1.2 g cm^{-3} and composed of 43% sand, 44% silt and 13% clay. After allowing six months for soil settling, orchard grass was planted on the lysimeters as the forage crop because of its hardiness and high nitrogen removal efficiency. Anaerobically-digested sewage sludge from Point Edward was applied in 1973 to the lysimeters until the sludge loading rate had reached 1500 kg TKN/ha, which is approximately ten times higher than the current field practice in Ontario province.

The fate and movement of petroleum hydrocarbons in the silt loam was determined by taking core samples from the lysimeters one year after sludge application. The individual core was sub-sectioned into 0-1, 3-4, 6-7, 11-12, and 19-20 cms to form various depth fractions. Two grams of soil (wet wt) were added to a 50 ml glass centrifuge tube, followed by the addition of 10 ml of chloroform-methanol mixture (3:1 v/v). The centrifuge tube was capped with a teflon-lined screw cap and the contents were mixed vigorously for one min. using a vortex mixer. After centrifuging the contents at $4000 \times g$ for 10 min, the chloroform extract (bottom layer) was transferred to a separatory funnel using a pasteur pipette. The soil layer was extracted again with 5 ml of the above solvent mixture and then 30 ml of distilled water was added to the pooled extract to produce two phases. The chloroform extract was finally concentrated to 1 ml with a stream of nitrogen.

The separation and determination of petroleum hydrocarbons were achieved using a Beckman GC-65 gas chromatograph, equipped with a dual flame ionization detector. The dual $180 \times 0.63 \text{ cm}$ stainless steel columns were packed with 2% OV-1 on 80-100 mesh Chromosorb W-AW-DMCS. The instrument was programmed as follows: linear temperature programme from 60 to 300°C at 5°C/min , followed by isothermal conditions at 300°C for 30 min; injection port, 275°C ; detector, 325°C ; helium, 60 ml/min; hydrogen, 45 ml/min; and air, 300 ml/min.

RESULTS AND DISCUSSION

The successive application of sewage sludge (March 28, June 19 and September 8, 1973 at the rate of 500 kg TKN/ha each time) to the lysimeters has resulted in an accumulation of high concentration of petroleum hydrocarbons (113 mg g^{-1} soil dry weight) in the top 0-1 cm soil layer in 1973. The chromatographic data (Figure 1) implied that petroleum hydrocarbons were a significant portion of the chloroform extract of this top 0-1 cm soil section from the lysimeters. This finding was further substantiated by field observation that some phytotoxicity (yellowing of leaf tips) occurred to the growing orchard grass in the lysimeters. The toxicity of hydrocarbons to various plants has been reported by Baker (1970). However, during the 1974 growth season, this toxicity effect was not observed and analysis of soil cores from the lysimeters taken on September 5, 1974 (362 days after application) revealed that substantial amounts (77%) of the petroleum hydrocarbons in the top 0-1 cm soil fraction had been degraded. This would probably explain the disappearance of the phytotoxicity effect to the growing crops in the lysimeters.

Figure 2 shows the gas chromatograms of the chloroform extract of the top 0-1 and 6-7 cm core sections one year after the last sludge application. In these and other chromatograms, selected peaks were identified by the use of internal and external standards in separate runs. Normal alkanes identified in this way were marked with carbon numbers in the figures. When comparing these chromatograms, attention should be paid to both quantitative and qualitative aspects. For example, the profile of the gas chromatogram of the 6-7 cm core fraction (Figure 2b), taken on September 5, 1974, resembled the 1973 0-1 cm soil sample (Figure 1) more than that (Figure 2a) of 1974. The reason for that was probably due to the anaerobic conditions prevailing with soil depth which tends to retard the microbial degradation of hydrocarbons. It has been known for some time that hydrocarbon biodegradation requires an aerobic environment (McKENNA and KALLIO 1965). Judging from the pattern of g.c. changes, it is apparent that the n-alkanes which represent the bulk of petroleum hydrocarbons on the lysimeters have been extensively degraded after one year land disposal. This is understandable as n-alkanes are normally less persistent towards biodegradation when compared with other petroleum fractions (HUMPHREY 1967).

The vertical distribution of sludge petroleum hydrocarbons in silt loam system after one year's land disposal is shown in Figure 3. The rapid decrease of hydrocarbon concentration with soil depth from 26.1 mg g^{-1} soil at 0-1 cm to 1.8 mg at 3-4 cm and 0.7 mg at 6-7 cm would suggest that the silt loam has an enormous capability to degrade as well as to retain the lipophilic substances such as hydrocarbons. Addition of sludge to soil would also enhance the soil's biological activity (VARANKA et al. 1976) which should, in turn, accelerate the degradation of hydrocarbons in soil.

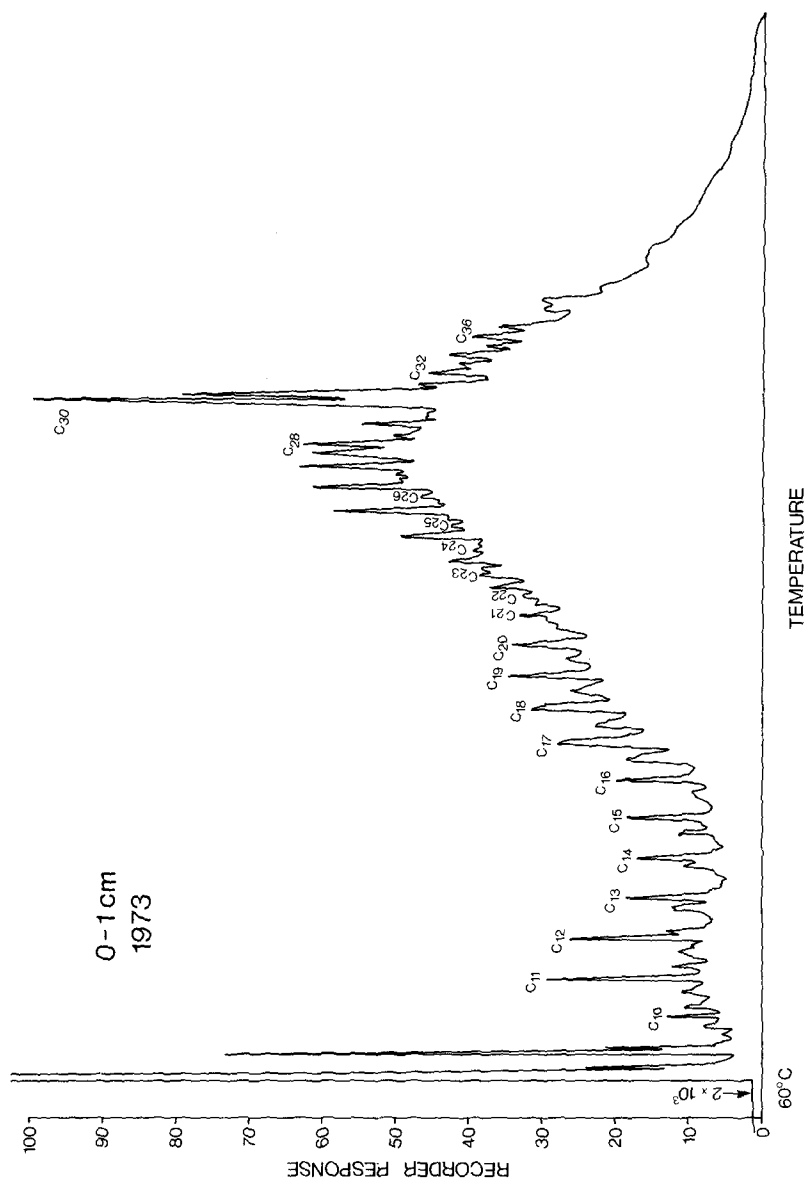


Figure 1. Gas chromatogram of the chloroform extract from the 0-1 cm core section sampled on September 12, 1973.

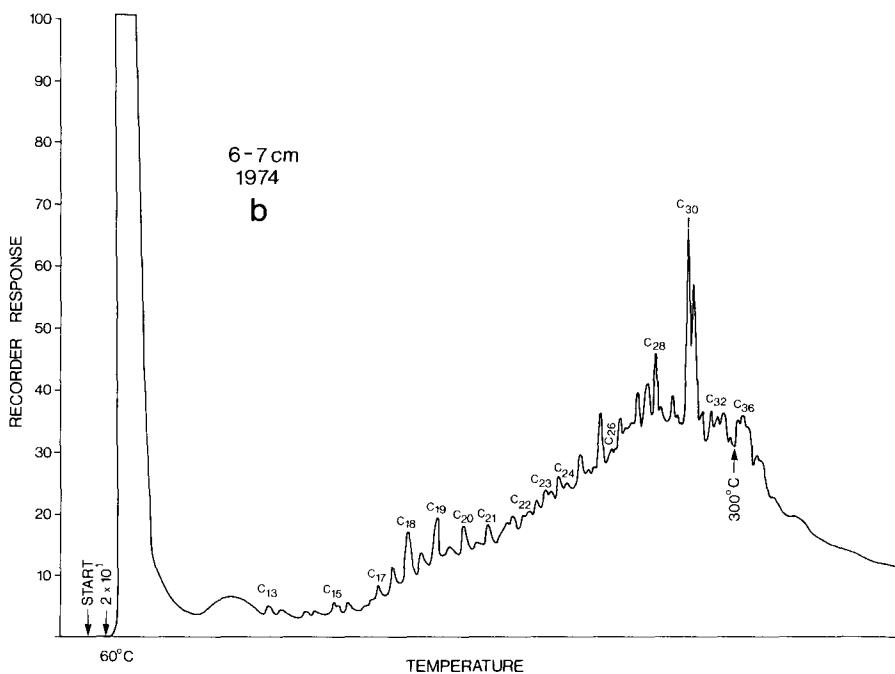
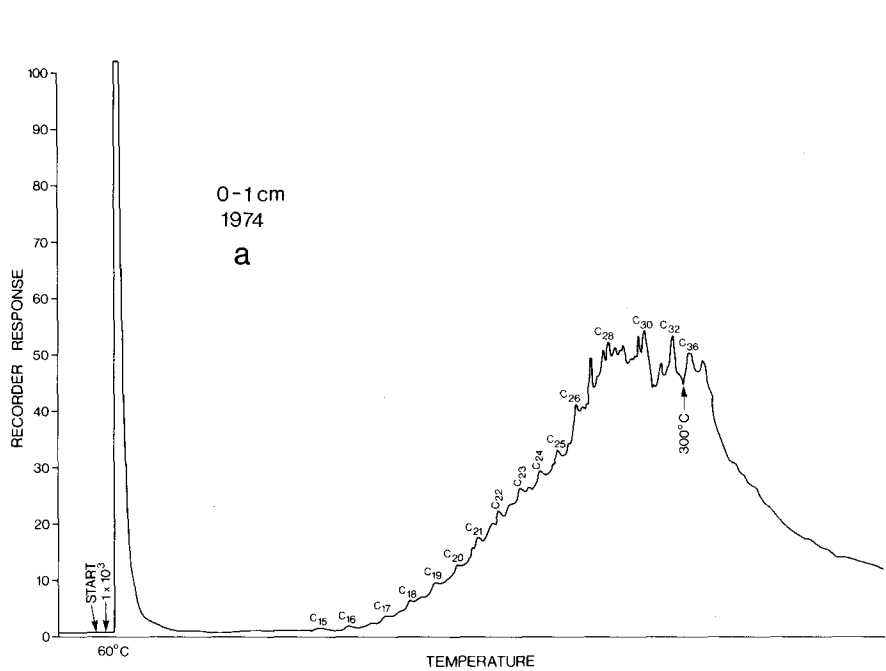


Figure 2. Gas chromatograms of the chloroform extract from the 0-1 cm (a) and 6-7 cm (b) core sections sampled on September 5, 1974

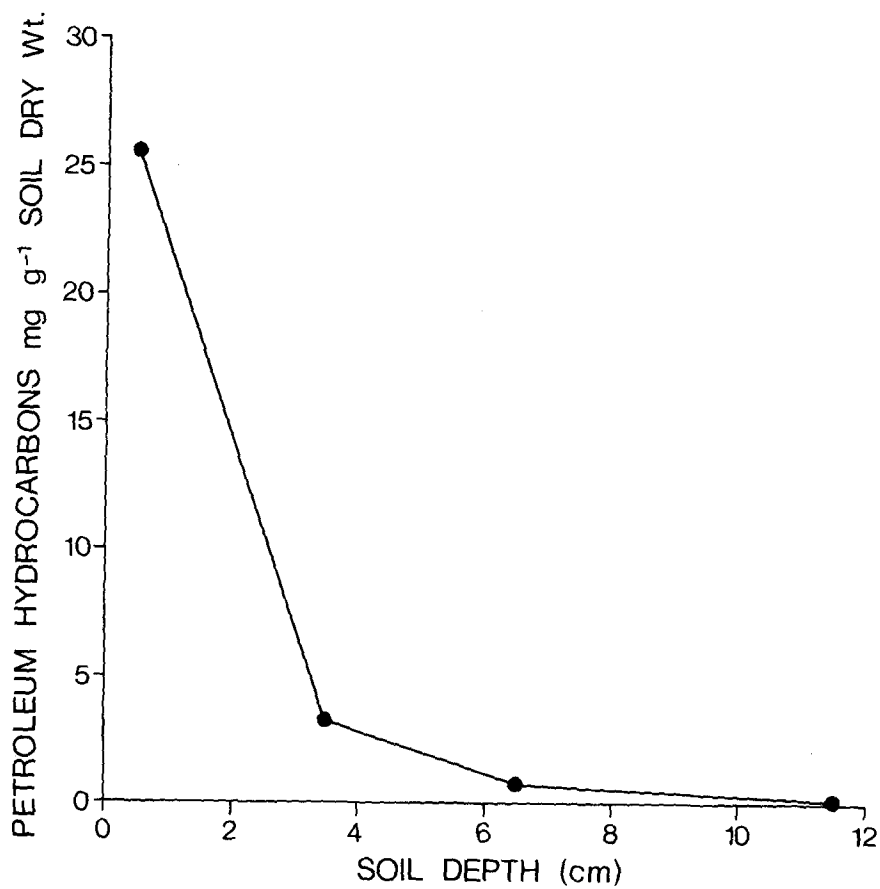


Figure 3. Vertical distribution of sludge petroleum hydrocarbons in silt loam after one year land disposal.

Normal sewage sludge does not have such high concentrations of petroleum hydrocarbons as that from the Point Edward sewage treatment plant (LIU 1978). Moreover, the sludge application rate used in the present study is at least 10 times higher than the current field practice in Ontario, therefore, there would be very little possibility of building up any significant level of petroleum hydrocarbons in the agricultural soil system by sludge land disposal. In view of the complexity of the problem of sludge disposal and the enormous economic and beneficial importance of sludge farmland disposal, further investigations, such as long-term study of the fate of other toxic organic compounds, for instance pesticides in sludge after land disposal, should also be carried out.

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