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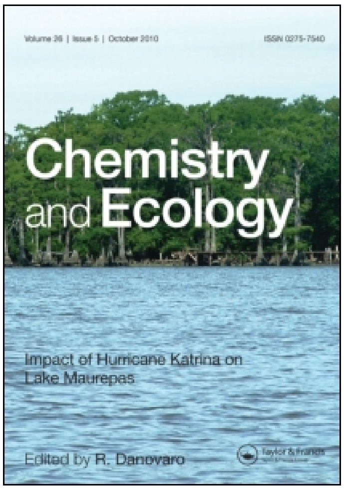

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A REVIEW OF WASTE TYRE UTILISATION IN THE MARINE ENVIRONMENT

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Waste tyres pose a major disposal problem on land creating a fire hazard and, in warmer climates, providing breeding pools for mosquitoes. The void space in tyres makes them unsuitable for land burial. Schemes to use shredded tyres for road bases and asphalt filler are being pursued in the USA. Tyre combustion for electricity production is being investigated in the UK.

The widespread availability and durability of tyres has led to their use in the marine environment for breakwaters/coastal defence structures and as artificial reefs for promoting fisheries.

Tyres have a low density and have been used in floating breakwaters. Schemes have been proposed to protect and strengthen shorelines with tyre structures.

The void space in tyres facilitates the construction of artificial reefs to attract fish. The most intensive use is in the south west Pacific and Australia. Tyre surfaces are colonised by algae and a wide range of faunal species, including corals and shellfish. The wide acceptance of tyres as a suitable reef construction material appears to be based largely on these observations. Experience of initial poor deployment practices in the USA led to tyres washing ashore after storms and resulted in the banning or restriction of their use in coastal states of the USA. A review of the scientific literature has yielded limited information on the environmental impact of tyres and in particular the leaching of heavy metals and organic compounds from tyres into sea water.

Preliminary results of tyre dust/sea water leaching studies are presented. These identify zinc as the major leachate (totalling 10 mg/tyre after 3 months). Diluted leachates have not shown significant effects of the growth of the phytoplankton *Phaeodactylum* and *Isocrysis* .

Further work to characterise the sea water leaching of tyre compounds is recommended.

KEY WORDS: tyres, waste utilization, zinc leaching, biological colonization

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INTRODUCTION

Millions of waste tyres are produced each year around the world. In the USA, Candle (1985) estimates that 200 million tyres accumulate annually. Whilst tyres can be retreaded to prolong their useful life they are eventually scrapped. Only a very small fraction of finely divided scrap tyre can be used in the manufacture of new tyres.

Tyres have been stockpiled in massive land dumps where they pose a serious fire risk. This is a world wide problem. Once alight they are often impossible to extinguish, generating vast quantities of smoke, toxic fumes, oils and liquids. In warmer climates there is an additional disease risk from mosquitoes breeding in rain-water pools in tyre rims (Dorer, 1978). The open structure, and hence bulk of tyres, has led to the European Union calling for a ban on their use in land waste burial sites. Incineration of tyres to produce energy is being instigated in a pilot project in the UK. The operators of this plant will only use tyres direct from a known source and will not consider those which have been stockpiled because of uncertainty over potential contamination. In the USA shredded tyres have been used for road pavements and mixed with asphalt (Serumgard and Blumenthal, 1993). However, in the UK, the economic viability of shredding tyres has been reduced following the availability of subsidised tyre shred from Germany.

UTILISATION IN THE MARINE ENVIRONMENT

Tyres are a familiar sight in ports, where they are commonly used as dock and boat fenders. The other main uses in the aquatic environment for tyres are for coastal protection and artificial reefs, both functions reflecting the resilience and availability of scrap tyres.

Breakwaters/Coastal Defence

Two extensive artificial reef bibliographies (Stanton *et al.*, 1985 and Berger, 1993) list some 60 tyre breakwater papers and over 200 references to tyre-built artificial reefs. The buoyancy of tyres has been utilised in the construction of floating breakwaters to absorb wave energy for shore and harbour protection (Armstrong, 1978) or protection of floating fish farms (McGregor, 1978). Bishop (1980) has produced a manual for the design and construction of floating tyre breakwaters.

Tyres have been used onshore to reinforce and protect the shoreline from erosion by storm wave action. In the U.K., an embankment for coastal protection has been built in Inverness using scrap tyres (Searle, 1985). There is also a proposal to use concrete filled compressed tyre bales to construct submerged offshore breakwaters for defence of the Holderness coast, NE U.K.

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Artificial Reefs

As with coastal defence applications, the resilience and availability of tyres has made them popular for building artificial reefs for fishery enhancement both in fresh and sea water. The open shape which causes problems for land disposal is a positive advantage when creating structures with multiple niches and high profile. Stanton *et al.* (1985) and Berger (1993) describe tyre reefs from North America, Caribbean, Europe, Middle East, Asia/Pacific and Australia.

Tyre artificial reefs are particularly popular in the south west Pacific (Philippines and Malaya). In Malaya, 54 reefs have been created using 1.5 million tyres (Zakaria, 1993). Stone *et al.* (1975) lists some 40 east US coast reefs which in total have utilised 700,000 tyres. A subsequent review of artificial reefs along the Atlantic seaboard of the USA (McGurrin, 1988) lists 73 tyre reefs. Those deployed in New Jersey alone, between 1984 and 1987, incorporated 7000 cubic yards of tyres. In Australia there are some 30 tyre artificial reefs (Kerr, 1992) making this the most frequently used reef material. At least 260,000 tyres have been used in their construction.

The buoyancy of the tyres requires that structures be well secured and ballasted. In the USA initial attempts to construct tyre reefs were poorly engineered and the tyres were quickly washed ashore by storms. This led to restrictions on their use or banning in some States (e.g. California and Washington) (Stone, 1985). McGurrin's (1988) review of Atlantic reefs describes the successful concrete ballasting of tyre units employed in different programmes in various States.

The majority of papers concerning tyre artificial reefs concentrate on the fish populations and catches. Tyre reefs have also been shown to be utilised by lobsters (Hruby (1979) off Massachussets; Briggs and Zwacki (1974) off New York). However, when considering the possible environmental impact of tyres in the marine environment, the growth of organisms on the surface of tyres is more significant than the mobile fauna, as their exposure to any chemical release is greater.

Colonisation of tyre surfaces by a range of flora and fauna has been described by:

Chin and Simons (1991), British Columbia; Vyshkvartsev and Lebedev (1990) Japan Sea; Prince *et al.* (1986), Smith Mountain Lake, USA; Laufle (1982), Washington; Alfieri (1975); Dewees (1970), California; Zawacki (1971), New York; Gordon (1972), Florida; Anon (1969), France.

In a comparison of settlement on various materials, Laufle (1982), noted preferential settlement of hydroids on tyres.

Algal colonisation of tyres has been described by:

Reimers and Branden (1991), Australia; Buckley (1982), Washington; Tsuda and Kami (1973), Guam; Matthews (1966), Florida.

In the Philippines there has been extensive loss of natural coral reefs (pers. com. H. Yap) through damaging fishing methods such as trawling and the use of explosives and removal of coral for curios and building materials. Tyre artificial reefs have been deployed to restore the former habitat diversity. There are studies of coral growing on such tyre reefs (Alcala *et al.*, 1981 and Gomez *et al.*, 1982). Fitzhardinge and Bailey-Brock (1989) compared the growth of corals on tyres, concrete and metal in Hawaii; the latter two were found to be more effective substrates. One problem with tyres is that they can flex during storms and thus shed rigid epifauna.

Tyres have been shown to make suitable surfaces for oyster settlement and growth. Currently, in Jamaica, commercial oyster (*Crassostrea rhizophorae*) culture spat are collected on tyre culches, with 6 inch square plates of cut tyre spaced along ropes (pers. com. M. Jones Williams). Tyre rims have been used as oyster culch (Anon, 1967). Querellou *et al.* (1983) describe a mixed reef of tyres and concrete blocks off Palavas, Montpellier, France which performed well as an oyster spat collector.

The US National Arificial Reef Plan (Stone, 1985) includes tyres as a reef construction material noting that no toxic effects attributable to leaching or decomposition have been demonstrated.

Although there are few descriptions of compounds leaching from tyres in the sea (described later) there has been unsubstantiated speculation about possible effects. A Greenpeace study (Vallette and Spalding, 1990) describes a case of US waste tyre dumping in the Marshall Islands (Pacific) supposedly to create artificial reefs. One of the many criticisms of this practice was the claim that the rubber contains toxic substances that inhibit marine encrusting. In a recent MAFF (1993) review of coastal defence and the environment there is a suggestion that tyres have been shown to be toxic to crustacea, though no references are given.

REVIEW OF LEACHING STUDIES

Tyres are manufactured from a wide range of chemical compounds which vary between type, manufacturer, date of production and country of origin of the tyre. Within a tyre there are different zones such as the tread, undertread, wall and liner, which are required to possess different physical properties. Both natural rubber and synthetic polymers (e.g. butadiene) are used in the production of a single tyre. Carbon black is used in large quantities as a reinforcing agent and filler. Oils are used to aid mixing of the components and to modify physical properties. Vulcanising agents such as sulphur, zinc oxide and organic peroxides promote polymerisation reactions at elevated temperature during manufacture. Accelerators (e.g. thiazole compounds)

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and retarders are used to control the rate of these reactions. To extend the life of the rubber, antidegradants are used. The British Rubber Manufacturers' Association code of practice on the toxicity and safe handling of rubber chemicals lists more than 100 compounds. Most of these are chemically transformed during the manufacturing process thus making it difficult to predict the leaching from waste tyres.

There is little published information about the leaching of compounds from tyres in sea or fresh water. The references include a number of unpublished reports.

Nozaka *et al.* (1973) concluded that no harmful substances were leached from a cut sample of used tyre soaked in fresh water. Details in this work are limited.

Mayo *et al.* (1974) undertook laboratory studies with tyre dust shaken with both fresh and salt water. This concluded that the principal component lost from tyres was zinc, equivalent to 20 lb of metal per century from 1 million tyres (about 10 mg/tyre). The organic material lost was studied by thin layer chromatography followed by infra red absorption. The presence of an aromatic carboxylic acid was inferred from this data.

An indirect study of tyre leaching was undertaken by Stone *et al.* (1975) using fish kept in tanks with tyres. The pinfish (*Lagodon rhomboides*) and black sea bass (*Centroprisis striata*) were chosen as both were resident on tyre reefs. Six used tyres were placed in 2000 l fibreglass tanks with a constant sea water flow-through. The supply rate used (15 l/min) was equivalent to renewal every two hours. At time intervals up to 100 days the fish tissue was analyzed for zinc, PCBs and organochlorine pesticides. The authors acknowledge that neither of the latter types of chemicals are used in manufacture but suggest the possibility that waste tyres could be subsequently contaminated. No significant differences were found between the fish in tanks with and without tyres. Given the high sea water turnover rate, it may have been difficult to detect any effects due to leaching. However, this approximates to a real situation rather than a worst case study.

Australia has made a massive investment in tyre artificial reefs. The State of Victoria Environmental Protection Agency commissioned a study employing salt water elutriation of cut used-tyre pieces (Probe Analytical, 1990). No release of polycyclic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) or heavy metals was detected. This flow-through test could be criticised for limited water/tyre contact time and area, but as with the Stone *et al.* (1975) study, it reflects realistic exposure conditions.

The Rubber Manufacturers Association in USA commissioned a study of a wide spectrum of rubber products, including tyres. This examined the leaching of heavy metals (excluding zinc) and release of volatile organics (including methyl ethyl ketone, carbon disulphide, toluene and phenol). The conclusion reached was that none exceeded the levels detailed in the EPA's toxicity characterisation leaching procedure. The exclusion of zinc and the limited range of organic compounds make these results of limited value to tyre studies.

There is considerable interest in the use of shredded tyre in road construction both as a base and when mixed with asphalt. The Minnesota Pollution Control Agency commissioned leaching studies (Twin City Testing Corp., 1990) of shredded tyre material to determine its suitability for roadway support.

One attraction is that it is considerably lighter than conventional rock rubble and would be useful over waterlogged ground. Cut tyre samples were leached in a range of solutions from pH 3.5 to pH 8. In acidic conditions, heavy metals (barium, cadmium, chromium, lead, selenium and zinc) were released in greater quantities. Conversely, total PAH and petroleum hydrocarbon release was greater under basic conditions. The release of organics from the tyre shreds was similar to that from asphalt.

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A more recent study by Eldin and Senouci (1992) involved an engineering field trial of a road pavement using tyre chips. The runoff/leachate from this site was collected and analyzed for a range of metals. Comparison with statutory limits leads to the conclusion that there was little likelihood of environmental damage. In the absence of control data, it is difficult to determine the actual effect attributable to leaching from the tyres.

A series of similar studies (Kellough, 1991; B.A.R. Environmental Inc., 1992; Goudey and Barton, 1992) have been undertaken recently in Canada to examine the leaching from tyres in fresh water. All three studies used glass aquaria with fresh water and cut tyre pieces. Samples of the water were removed at intervals and test species were kept in the water. All groups found that these leachates were toxic to rainbow trout (*Oncorhynchus mykiss*). However, B.A.R. Environmental Inc. found that leachate from used breakwater tyres was not acutely toxic to the trout, indicating the loss of active compounds with time. Kellough and B.A.R. Environmental Inc. found that the water flea, *Daphnia magna,* was unaffected by any tyre leachates. However, Goudley and Barton using much higher tyre/water ratios, found toxic effects in this species. B.A.R. Environmental Inc. found no effect on fathead minnows. Kellough measured levels of heavy metals, PCBs, organochlorines and PAHs in the water and exposed fish tissue. Of the metals, only zinc was detected in elevated concentrations in the water samples. PCBs and organochlorines were not detected in any samples. The PAHs anthracene and chrysene were detected in control, test water and fish samples with no significant difference.

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EXPERIMENTAL STUDIES

Tyre material was used to examine the leaching of compounds into sea water. The tyres were finely divided to provide a large surface area giving increased leaching rates and leachate concentrations and thus making detection easier. This work was designed as a 'worst case' study with very high surface to volume ratios. Two sources of finely divided material were used; tyre crumb and tyre dust. The tyre crumb (supplied by Pirelli Ltd.) is derived from whole scrap tyres and had been passed through a 420 μm sieve. Particle size of a sample was analyzed under a light microscope to determine the effective surface area (800 cm^2/g). The dust from a tyre retreaded (Motorway Remoulds Ltd.) contained a wide range of particle sizes which were sorted into different categories; 2000, 1000, 500 and 250 μm using Christison steel sieves.

Sea water for the study was taken from Poole Bay off the Dorset coast, away from land pollution sources. It was collected in cleaned 25 l polyethylene carboys and then filtered through 1 μm Whatman GF/C filters. Typically, leaching studies with dust or crumb used 33 g of material shaken with 750 ml sea water. This mass of crumb was calculated to have the surface area equivalent to 7 tyres per litre. Ground glass stoppered bottles (for organic studies) (1l) or screw top polythene containers (for metal studies) were used. All containers were cleaned with 'Micro' detergent, then rinsed with acetone followed by soaking in 5% hydrochloric acid. Final rinsing was with 'Milli-Q' Millipore deionised water. Samples were shaken continuously in a circular motion at 100 rpm for up to 3 months. The sea water dust/crumb mixture was prefiltered through 64 μm plankton netting before final filtration through 1 μm 4.25 cm Whatman GF/C filters.

In all the dust or crumb leaching studies a decrease in the order of 1 pH unit was observed.

Metal Studies

Heavy metal (cadmium, copper, chromium, lead, nickel and zinc) analysis was by flame atomic absorbtion spectrometry using a Pye Unicam SP9 AAS. Leachate samples (5 ml) were acidified with concentrated nitric acid (1 ml). BDH Spectrosol standard solutions were used to prepare calibration curves with each batch of analyses. The accuracy of these was checked by nitric acid digestion and analysis of a certified reference material, lobster hepatopancreas (Tort-1, National Research Council Canada, Division of Chemistry, Marine Analytical Chemistry Standards Programme). Results for the diluted standard solutions were within one standard deviation of the certified reference values.

The only heavy metal detected in the sea water leachates was zinc, other metal concentrations were below detection limits (Cd–0.01 μg/l; Cu,Cr,Ni,Pb–0.05 μg/l).

Two experiments were undertaken to demonstrate the relationship of zinc release to particle surface area. In the first (Figure 1) different quantities of crumb were used. Leachate concentration was directly related to quantity of crumb. A second experiment used the different size fractions from the retreaded dust (Figure 2). The doubling of the size fraction from 250 to 500 μm resulted in a halving of the release rate corresponding to halving the surface area of tyre material.

The 33 g of crumb was calculated to be equivalent to 5 tyres, thus the final concentration in 750 ml sea water (Figure 1) corresponds to a release of 13 mg per tyre. Leaching of zinc from 4 whole scrap tyres in separate tanks of 25 l sea water was also monitored. The increase in concentration of zinc above the blank was in the order of 0.4 μg ml. This equates to a loss of 10 mg per tyre, similar to the value calculated from tyre crumb results above and by Mayo *et al.* (1974).

Large quantities (1–2%) of zinc oxide are used in manufacture to assist the vulcanisation process. Acid digestion of a tyre crumb sample showed 1% zinc by weight. However, loss takes place only from the outer surface and only 10 mg of a potential 100 g appears to be leached. Presumably as the tyre fabric degrades further

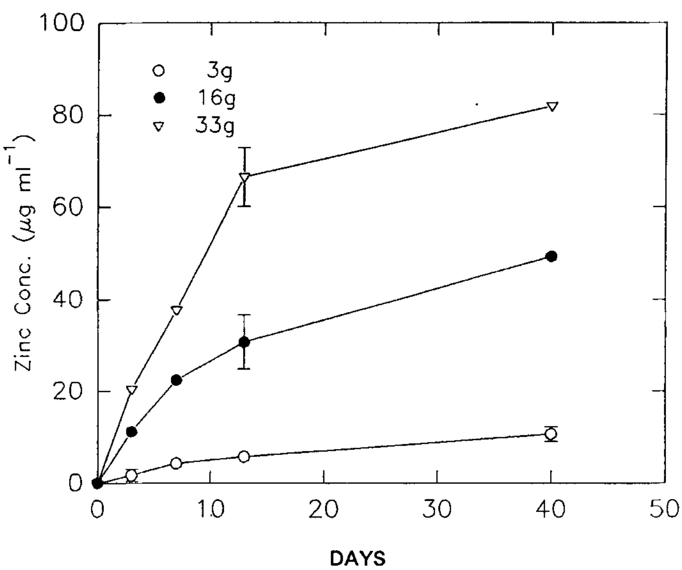

Figure 1 Effect of mass of tyre crumb (in 750 ml sea water) on zinc leachate concentration, showing mean and standard deviation.

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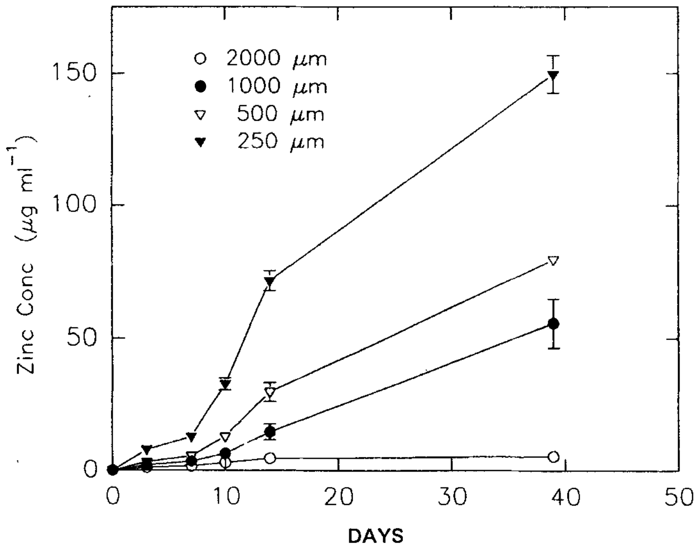

Figure 2 Effect of tyre dust (33 g/750 ml sea water) particle size on zinc leachate concentration, showing mean and standard deviation.

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release would occur. However, a tyre below the surface of the sea is not exposed to photodegradation and is maintained in a constant pH8 medium. This can be contrasted with greater loss of zinc and measurable loss of other metals under acidic conditions such as may be found in dump-sites on land. Greater loss of heavy metals under acidic conditions was reported by the Twin City Testing Corp. (1990).

Zinc is present in sea water and is an essential element for the growth of many organisms such as crustaceans and fish (Bryan, 1976). However, high concentrations can inhibit growth, e.g. 15–20 $\mu g\ l^{-1}$ reduces the growth of phytoplankton (Hollibaugh *et al.*, 1980). The amount of zinc observed in marine tissues is often much greater than the amount required to satisfy the needs of the enzyme systems (Coombs, 1972) and hence there are protection mechanisms operating against the inhibitory effects of high concentrations. This is achieved by a variety of mechanisms including binding with proteins and formation of metal granules. Oysters often contain high levels of zinc; Bryan (1976) quotes normal concentrations for *Ostrea edulis* in excess of 4000 ppm dry wt for 'normal' areas.

Organic Studies

A rapid means of assessing the organic content of leachate was by fluorescence scanning. Many aromatic compounds with multiple benzene rings fluoresce. Filtered samples in a 10 mm quartz cuvette were scanned using Perkin-Elmer LS-5 Programmable Fluorescence Spectrometer Detector. Instrument and data acquisition were controlled by a PC running Perkin-Elmer Computerised Luminescence Spectroscopy (PECLS) software. The excitation wavelength was started from 230 nm whilst emission fluorescence was repeatedly measured across the range of 250–600 nm. The spectra were quantified using quinine sulphate standard solutions.

The results of fluorescence scans of crumb leachates after different leaching times are shown in Figure 3. The peaks indicate the presence of a range of single to multiple

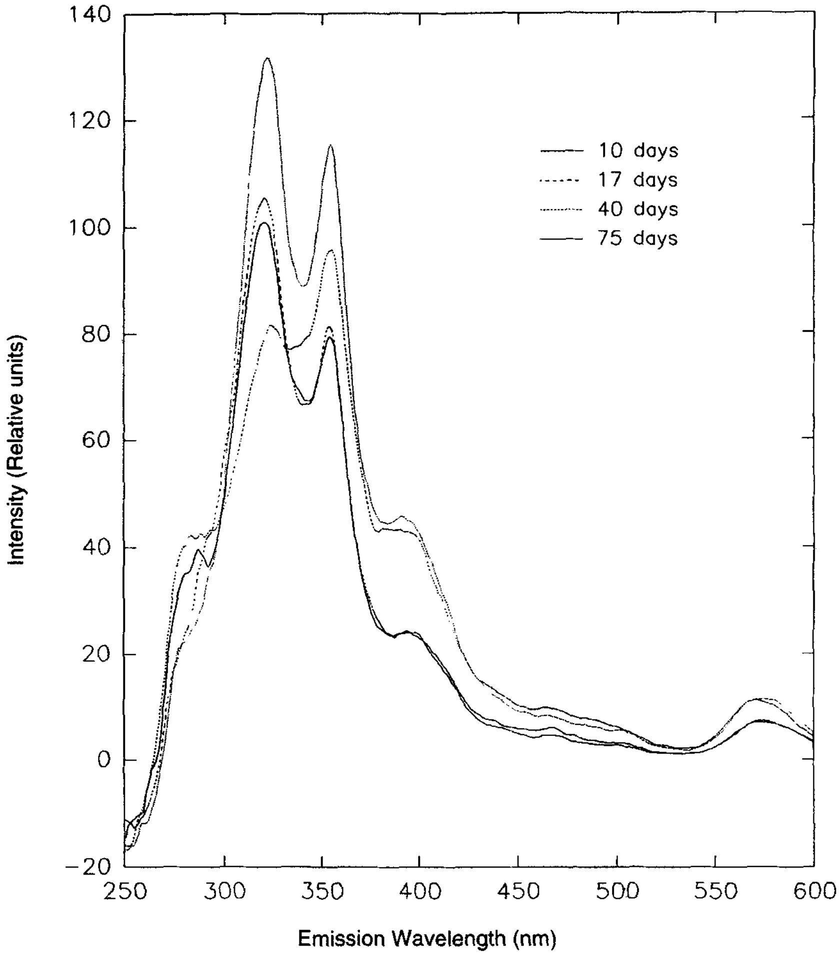

Figure 3 Synchronous fluorescence scans of tyre dust/sea water leachates.

benzene ring compounds. There is an increase in peak heights with exposure time. Total release after 75 days was estimated to be in the order of 5 mg/tyre.

This organic material has not been characterised but indicates the presence of a number of polyaromatic hydrocarbons (PAHs). These are likely to be derived from the process oils used in tyre manufacture. The decrease in pH of the leachates is probably due to the rosin acids used as surfactants in manufacture. Mayo *et al.* (1974) found evidence of carboxylic acids in tyre leachates.

Bioassays

Another approach to studying leachates is to test their effects on the rate of growth and/or survival or organisms. It is difficult to identify all the compounds present in leachates, and even if it were possible to determine the individual toxicity of each one, there may be cumulative effects.

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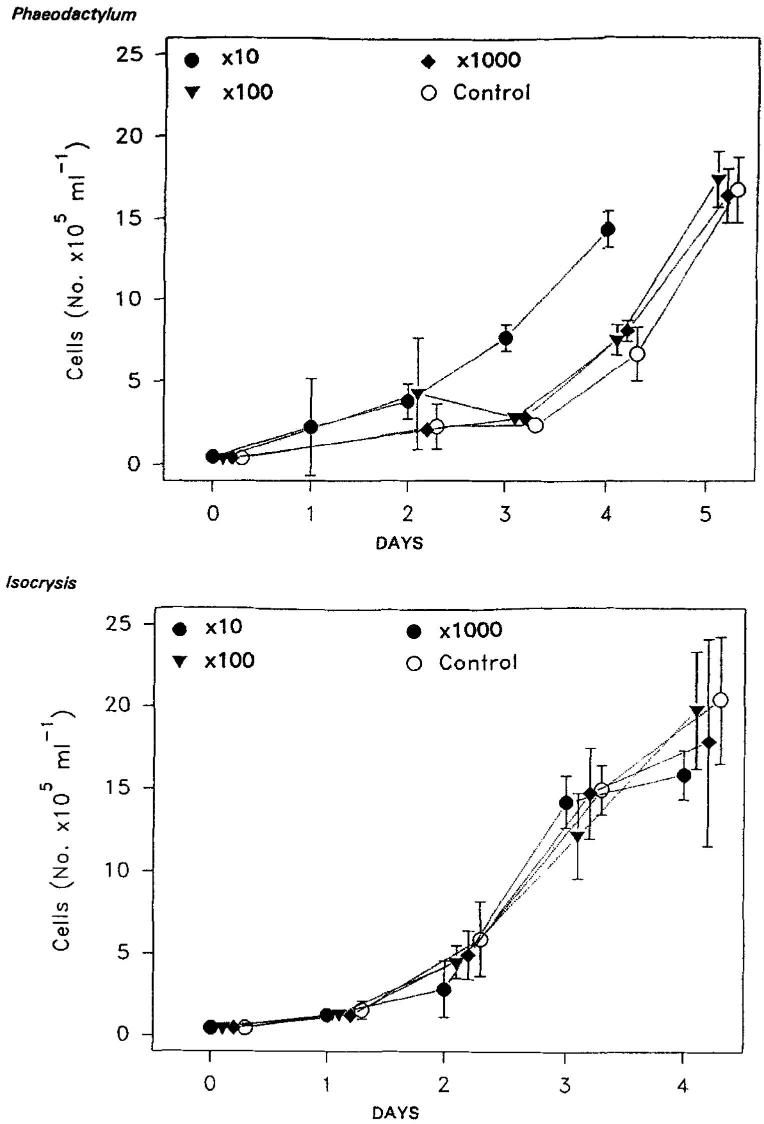

Figure 4 Growth of the phytoplankton *Phaeodactylum* and *Isocrysis* in different dilutions of tyre crumb leachates, showing mean and standard deviation.

Two species of phytoplankton were used: *Phaeodactylum tricornutum* and *Isocrysis galbana*. All experiments with leachates were run in triplicate along with sea water blank controls in autoclave 250 ml conical flasks. Sea water for dilution of leachates and controls was filtered through 50 μm plankton mesh, then Whatman 1 μm GF/C filters and finally Whatman 0.45 μm membrane filters. It was then sterilised by autoclave. Leachates were filtered in the same way as the sea water before dilution to the required concentration making a total volume of 200 ml. These diluted leachates were chemically sterilised as described above. Nutrient and vitamin stock solutions

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were added to all flasks plus 2 ml *Phaeodactylum* or *Isocrysis* stock culture. The flasks were stoppered with muslin covered cotton wool plugs and incubated at 20°C under fluorescent lighting. Daily cell counts from each flask were made using a Coulter Counter (model PCA1). Three subsamples (0.05–1 ml) from each flask were diluted with 30 ml Isoton II. The Coulter counter was initially checked with uncontaminated algal cultures to determine the appropriate channels (Nos. 2–5, 3.17–6.35 μm).

The tyre crumb leachates represent very high tyre to water ratios (7 tyres/l) so bioassays were carried out at a number of dilutions (×10, ×100 and ×1000). The effects which tyre crumb leachates (after two weeks shaking) have on *Phaeodactylum* and *Isocrysis* are shown in Figure 4.

There does not appear to be suppression of growth rate at any of these dilutions.

CONCLUSIONS

Millions of waste tyres are in use around the world for artificial reefs and breakwater/coastal defence applications. Waste tyre artificial reefs have been shown to be successful in attracting fish. More significantly, for assessment of environmental impact, numerous studies have described the successful colonisation of the tyre surfaces by algae and a wide range of faunal species including corals and shellfish. The wide acceptance of tyres (including developed countries such as the USA and Australia) as a suitable reef construction material appears to be largely based on these observations.

A few studies have examined the leaching of chemical compounds from tyres. Most of these have been concerned with leaching in fresh water. In the US release of heavy metals and organic compounds from shredded tyre for road pavement has been studied. The effect of tyres in fresh water on test organisms has been studied recently in Canada. These studies have found leachates from recently immersed tyres to be toxic to trout but tyres which had been submerged for a number of years did not show any toxicity. Work with leaching in sea water has been limited in approach and scope.

In this study preliminary work has been undertaken with leaching of finely divided tyre material in sea water. Of the heavy metals (Cd, Cu, Cr, Pb, Ni, Zn) analyzed in the sea water leachates only zinc has been detected. Zinc compounds are used in tyre manufacture (1–2% by weight). However, release has been shown to be from the outer surface only and total release estimated to be in the order of 10 mg/tyre (after 3 months leaching). This metal occurs naturally in sea water and is an essential element for many marine organisms. In an acidic environment, such as would be found on land, other heavy metals could be released. Leaching of organic material has been shown. Whilst this has not been characterised, the presence of PAHs and organic acids are inferred. Dilutions of the tyre crumb leachates appear not to have significant effects on the growth rates of the phytoplankton *Phaeodactylum* and *Isocrysis* .

Given the widespread use of tyres in the marine environment and current concern about the use of waste materials in the sea, the lack of published information about tyre leachates is surprising. There is a need to characterise more fully the leaching of compounds from tyres in the sea. Further bioassay work with a range of marine organisms would assist in determining the potential effects of these leachates. Another avenue of research would be analysis of organisms growing on tyre reefs to determine if there is evidence for bioaccumulation of the leached tyre compounds.

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