Environmental Effects of Terpenoid Chemicals: A Review[1]

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ABSTRACT

Entry of terpenes into the atmosphere occurs by volatilization of hydrocarbons from foliage, especially over forest lands. These compounds react photochemically with ozone, causing a blue haze. The total volume of reactive hydrocarbons released naturally appears several times greater than that from man's activities. Diterpenes also enter the environment through surface water runoff. Recently dehydroabietic acid and other terpenoidal compounds were found in ocean sediments, deposited by rivers. Few cases of air pollution can be related to industrial operations or processing of terpenes. Veneer dryer stack gas emissions cause an aesthetic problem, controllable by better modes of operation. Workers in soldering operations require protection from fumes caused by use of rosin cores. Rosin and rosin derivatives are virtually nontoxic to mammals and find use in food packaging applications. Sulfate turpentine is the source of pinenes for manufacture of synthetic flavors and perfumes. Resin acids and related diterpenes are, however, acutely toxic to aquatic organisms and can cause severe problems to fisheries if released in pulp mill wastes. Both monoterpenes and diterpenes are biodegradable. The future widespread use of toxaphene as an insecticide is uncertain because of its toxicity to aquatic fauna and relatively long persistence in soils and sediments. Recent results have demonstrated microbiological uptake of toxaphene components leading to problems of bioaccumulation through the food chain.

INTRODUCTION

Terpenes are naturally occurring chemicals found in plants and animals. The monoterpenes, which contain ten carbon atoms, are found in the essential oils of plants and are recovered as turpentine from coniferous softwoods or by the extraction of various annual plants. The diterpenes contain twenty carbon atoms and occur in conifers as resin acids and related alcohols and aldehydes. They are recovered as tall oil during kraft pulping or as resinous extracts or exudates from trees. Current research in natural product chemistry is discovering many diterpenes of novel structure in the tissues of marine plants and animals. Despite the widespread importance of the terpenes in the perfumery, naval stores, and chemical manufacturing industries, the effects of their release to the environment have not been fully reviewed (1).

In this article, the more general term "terpenoids" is preferred because it includes products, such as toxaphene, manufactured from natural terpenes.

NATURAL ESCAPEMENT FROM FOREST LANDS

Some years ago, Rasmussen (2) discovered that air in contact with plant foliage of five common tree species contained isoprene, not previously considered a natural product. The substance was identified by infrared and mass spectrometric analyses, and concentrations were measured in the range 0.5 to 24 ppb in air overlying mango foliage in Panama. Monoterpene hydrocarbons such as α- and β-pinene and limonene are emitted by conifer foliage at rates in the range 0.4 to 3.5 ppb/g/min/l dependent on rate of transpiration, structural integrity of the oil cells and resin glands, and foliage temperature (3). The emission of isoprene by hardwood foliage is light dependent and ranges between 0.05 and 2.4 ppb/cm^2/min/l for oak, cottonwood, and eucalyptus foliage. Some species – notably various spruces, sweetgum, yellow and balsam poplars – emit both isoprene and α-pinene. The annual global contribution of forest hydrocarbon emissions is estimated as 175 x 10^6 tons compared to 27 x 10^6 tons from all man's activities. According to Rasmussen's calculations, there is a sixfold greater emission level from natural than from man-made sources. Such emissions cause the blue haze that is so typical in forested areas on a hot summer day and have given rise to names of mountain ranges such as the Blue Ridge Mountains, the Great Smoky Mountains, or the Blue Alps. This blue haze is caused by photochemical reactions between terpene hydrocarbons and ozone.

Resin acids and other terpenes can enter the aquatic ecosystem through natural circumstances. During spring freshets, river levels rise and inundate swamps, or coniferous trees fall into the water as river banks are washed away. Although resin acids are very sparingly soluble in water, dehydroabietic acid has a significant solubility due to the influence of the aromatic ring. Recently, Simoneit (4) has found diterpenoids with the abietane skeleton (mainly dehydroabietic acid) in sediments of the northeast Pacific, the Black Sea, and the North Atlantic. These resin-derived compounds correlated with terrigenous clay components and with the presence of pollen. They probably enter the ocean basins by river transport and are useful chemical indicators of the origins of ocean sediments.

EMISSIONS FROM PLYWOOD VENEER DRYERS

Recently, concern arose in the Willamette Valley in Oregon over the blue haze from stack gas emissions of veneer dryers (5,6). Emissions consisted of two types of hydrocarbons: (a) those that condense readily on cooling and (b) those that remain volatile. The latter type resembles emissions from growing vegetation, while the condensable hydrocarbons are responsible for a bluish opaque haze emitted from some veneer dryer stacks. This haze is due to volatilization of resin and fatty acids but does not contribute to photochemical smog. The amount of condensable hydrocarbons was related to species. In terms of 10,000 ft^2 of 0.375 in veneer, white fir yielded 0.5 pounds; Douglas-fir heart wood, 3.2 pounds; Douglas-fir sap wood, 4.6 pounds; and ponderosa pine, 8 pounds.

The volatile hydrocarbon fraction contained photochemically unreactive methane and ethane plus moderately active monoterpene hydrocarbons. An average plywood plant emits 2.7 pounds of volatile hydrocarbons per hour which is equivalent to the production rate from 250 acres of forest land.

Although there is no evidence that veneer dryer emissions are harmful (7), they are aesthetically displeasing, and, as a result, better modes of operation, such as improved control of damper settings, and engineering

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modifications have been introduced by the industry. Such measures have ameliorated the aesthetic problem. Recently, another solution has been suggested at the Western Forest Products Laboratory in Vancouver (8). Spraying an aqueous solution of borax on the veneer surface during high-temperature drying prevents surface inactivation, which gives rise to unsatisfactory glue-wood interface bonding. Coincidentally, quantitative gas chromatography showed that the addition of borax to Douglas-fir *(Pseudotsuga menziesii)* or white spruce *(Picea glauca)* wood resin, during heating at 180 C, substantially lowered the loss of resin and fatty acids. The addition of borax to Douglas-fir resin raised the incipient temperature of haze formation by 30 C, when heated with ozone. No opacity measurements of veneer dryer stack gases have been reported on full-scale trials with borax.

ENVIRONMENTAL CONTAMINANTS FROM NAVAL STORES PLANTS

Recently, the Environmental Protection Agency has promulgated wastewater discharge regulations for gum and wood chemicals manufacturing point sources (9). These regulations set limits for BOD_5, total suspended solids, and pH of emissions from the following industrial subcategories: char and charcoal briquets; gum rosin and turpentine; wood rosin, turpentine and pine oil; tall oil rosin; pitch and fatty acids; essential oils, and rosin-based derivatives.

An example of a naval stores processing problem and its solution was cited by Lawrence (10). After oleoresin extraction, an appreciable quantity of oxidized resin sometimes remains, causing a very black smoke when the extracted chips are combusted. The solution is to follow the hydrocarbon solvent extraction with a further extraction using either acetone or methyl isobutyl ketone. This greatly reduces the pollutants in the smoke and yields an additional 5-8% of dark, oxidized resin.

The pyrolysis products from rosin core solder cause an air pollution problem of direct relevance (11). The thermal decomposition products include acetone, methyl alcohol, methane, ethane, abietic acid and related diterpene acids. These by-products irritate the eyes, nose, and throat of exposed workers, and a threshold limit value (TLV) of 0.1 mg/m^3 in workroom air has been established.

In the control study, human volunteers were exposed to pyrolysis fumes of gum rosin (grade WW) in an environmental chamber. Concentrations of aldehydes, as formaldehyde, ranged from 0.04 to 0.2 mg/m^3. About 1 mg of aldehyde was produced per gram materials pyrolyzed. Eighty percent of the subjects reported moderate to severe irritation to the eyes, nose, or throat at concentrations above 0.12 mg/m^3. Adaptation occurred quickly, however, and after one or two minutes, most subjects could no longer detect the odor. On the basis of these tests and tests with laboratory animals, a TLV of 0.1 mg/m^3 aliphatic aldehydes, (as formaldehyde) has been adopted for industrial workroom environments.

Crude tall oil and sulfate turpentine are important feedstocks for the naval stores processing industry. Unfortunately, both contain significant loadings of malodorous organosulfur compounds, and sulfate turpentine is difficult to purify. Bald (12) has described analytical procedures for methyl mercaptan and dimethyl sulfide in sulfate turpentine. Hrutfiord and McCarthy (13) examined the composition of kraft blow gas condensates and turpentine samples and identified the above compounds plus dimethyl disulfide and 4-methyl-2-pentanone. Later studies at the University of Washington identified 2-heptanone and 3-methyl-2-butanone in sulfate turpentine (14). More recently dimethyl trisulfide and diethyl disulfide were found in kraft effluents (15).

The analysis of wastewater from tall oil rectification has been described. Compounds present included phenol, o-, m-, and p-cresol, 2,5-xylenol, 2,4-xylenol, guaiacol, and $C_{6\text{-}22}$ fatty acids (16). The potential microbial toxicity of wastewaters from the purification of tall oil and turpentine has been considered (17). Although there was no evidence of bactericidal activity associated with the spent acid from acidulation plants, it was recommended that this highly contaminated stream (COD 220,000 mg/l) not be channeled to biological treatment plants.

MAMMALIAN TOXICITY TESTS AND FOOD USES OF NAVAL STORES PRODUCTS

Various grades of rosin, modified rosin, and rosin derivatives have undergone mammalian toxicity tests (18). A series of such products was fed to rats, mice, and guinea pigs, and no significant oral toxicity was identified. A similar observation was made in tests on a sample of condensed resin from the inside wall of a veneer dryer stack (7). Mixed resin acids, recovered from kraft effluent, were tested for mutagenic activity using a strain of *Saccharomyces cerevisiae* (19). The tendency to induce gene conversion was low or nonexistent.

A radioactive tracer technique was used to study the migration of rosin components from sized paper to exposed foods (20). Tritiated samples of both pale and dark rosin were used in the preparation of both waxed and unwaxed samples of many common types of highly sized paper and paperboard. A wide variety of solid, semisolid, and liquid foods with both high and low fat contents was exposed to the radioactive paper, under realistic conditions of time, temperature, and surface contact. From measurements of radioactivity transfer, it was concluded that the migration of rosin components from sized paper to food was below 9 mg/kg. This maximum transfer would occur if the foodstuffs were packaged in paper and paperboard containing the highest practicable amount of rosin size. Rosin derivatives are now used in food packaging and in the manufacture of chewing gum (21).

Fractionation of sulfate turpentine yields α- and β-pinene used in the synthesis of numerous flavors and perfumes. This subject has been well reviewed by Derfer (22,23) and by Zinkel (21). Products thus manufactured include menthol, geraniol, citral, ionones, terpineol, camphor, isobornyl acetate, and various essential oils including lemon, lime, peppermint, spearmint, and nutmeg. Polyterpene resins, also made from pinenes, find application in chewing gum manufacture.

TOXAPHENE

Toxaphene has been cited as the most widely used insecticide in the United States on the basis of an annual consumption in 1972 of 58 million pounds (24). This preparation, made by the chlorination of camphene, controls insect pests on cotton, soybeans, corn, and other crops, besides livestock.

Analyses of soil, sediment, and water samples have indicated that toxaphene persists as long as nine years after application during fish eradication programs in lakes (25,26). In studies of toxaphene persistence, Hughes (27) investigated its bioaccumulation in the food chain in treated lakes and found that fishes and sediments selectively accumulated different fractions. Recently, Paris et al. (28) showed that bacteria, fungi, and algae in natural aquatic systems also accumulate selected fractions. The sorbed portion is the least water-soluble and is not the result of metabolism, since autoclaved samples of these cultures sorbed as much toxaphene as did the viable cells. Equilibrium levels of toxaphene were established in a few hours, and there was no evidence of degradation even after extended time periods. Since these microorganisms are subject to predation by higher trophic levels in the food chain, bioaccumulation occurs whether the toxaphene is adsorbed within the organisms or on the outer surfaces.

Several research groups have examined the toxicity of toxaphene to aquatic organisms. Thus, Chaiyarach et al. (29) measured its acute toxicity and that of three other biocides (carbaryl, propanil, and molinate) to mosquitofish *(Gambusia affinis)*, grass shrimp *(Palaemonetes kadiakensis)*, crayfish *(Procambarus simulans)*, and mactrid clam *(Rangia cuneata)*, all of which, except Rangia, are freshwater species. The latter is found in estuarine waters. The 96 hr TL_m values for toxaphene were 0.008 mg/l, 0.036 mg/l, 0.21 mg/l, and 460 mg/l, respectively. (Median tolerance limit value is a term synonymous with LC50 and is defined as the concentration by volume in which 50% of the test animals expire within the defined period.) The toxaphene was significantly more toxic than the other insecticides in freshwater but was the least toxic to Rangia, partly because this formulation is more unstable in saltwater than in freshwater. Another study examined the uptake of ^{36}Cl-toxaphene by mosquitofish (30). The toxicity syndrome of this animal exhibits five stages, and the residue level corresponding to each stage was measured. When the fish were exhibiting the first toxicity response (stage 2), over 90% of the mean fatal residue level had been sorbed. Excretion of toxaphene was not observed following an 8-hr exposure to 2 mg/l, and metabolic alteration during the experiment was minimal. Another study examined the inhibition of ATPase activity in catfish *(Ictalurus punctatus)* tissues resulting from exposure to toxaphene (31). Inhibition of the enzymes from brain, gill, and kidney was observed. Toxaphene presents difficulties in biochemical studies because it is a complex misture of up to 175 components, and the need to examine the effects of individual constituents has been expressed. Chemical isolation work as a preliminary to further biochemical studies has been reported (32). The effect of toxaphene and of a variety of chlorinated insecticides on beef heart mitochondrial electron transport (33) was examined. Toxaphene inhibited both NADH-oxidase and succinoxidase activity in vitro. However, it was uncertain whether this enzyme inhibitory activity was significant in the toxicology of the intact organism.

The metabolic fate of ^{36}Cl-toxaphene in rats has been investigated (34). Over 50% of an oral dose was excreted within 9 days with most of the excreted radioactivity in the aqueous phase as ionic chloride. Less than 10% was found in selected tissues and organs one day after administration. Finally, the fetal toxicity of toxaphene in rats and mice has been examined (35). In the rat a reduction in fetal weight and a decrease in the degree of skeletal ossification were observed. An incidence of encephaloceles was the only fetal effect observed in the mouse. In both species the dosages were sufficiently high to cause maternal toxicity.

IDENTIFICATION OF FISH TOXICANTS IN KRAFT EFFLUENTS

The toxicity of resin acids to fish has long been recognized (36). Fish kills and resin-like taste of resident fish populations in Scandinavian rivers were attributed to the release of these compounds from pulp mills. Hagman (37) found resin acids poisonous to fish in concentrations higher than 1 mg/l, and sulfur compounds in the waste liquor were shown poisonous in relatively low concentrations. Van Horn et al. (38) reported that the minimum lethal concentration of the sodium soaps of mixed resin acids was 1.0 mg/l to four species of freshwater minnows and 3.0 mg/l to the freshwater flea *(Daphnia pulex)*. Later Maenpaa et al. (39) reported the mixed resin acid sodium soaps toxic to Daphnia at 1 mg/l but found that the free resin acids were nontoxic. Leach and Thakore (40) have attributed the toxicity of a sample of unbleached kraft effluent (KME) to juvenile coho salmon *(Oncorhynchus kisutch)* as 80% due to resin acid soaps and 20% to unsaturated fatty acid soaps. Resin acid soaps were identified as toxicants at several mills in the Maritimes by Row and Cook (41), who also reported resin acid concentrations of 59 mg/l in groundwood mill and hardboard plant wastes. Abietic acid was the major resin acid and correlated closely to fish toxicity. These authors reported mixed resin acids toxic to juvenile Atlantic salmon at 2 mg/l. Rogers (42) identified mixed resin acids as mainly responsible for bioassay failures of biologically treated bleached kraft effluent (BKE) to meet permit requirements at a British Columbia mill. The free resin acids killed sockeye salmon *(O. nerka)* at 2.0 mg/l. Resin acids also accounted for 60-90% of the toxicity of mechanical pulping effluent from two Canadian mills (43). The wood supply comprised spruce (75-85%), pine (15-25%), and fir (5%). Here, the acids were in the free form and not as sodium soaps. The median lethal concentrations (LC50) values for various resin acids have been measured in 96-hr static bioassays with rainbow trout *(Salmo gairdneri)* (43). The values are: dehydroabietic 1.1; pimaric 0.8; abietic 0.7; palustric 0.5; and isopimaric 0.4 mg/l.

Other components of pulping wastes are also acutely toxic to fish. Thus the fatty acid fraction of tall oil soap killed minnows at 5.0 mg/l (38). Unsaturated fatty acid soaps, but not those of saturated acids, were minor toxicants in KME (40). The unsaponifiable fraction of lodgepole pine *(Pinus contorta)* wood extractives killed sockeye salmon (44); the toxic effect was more acute in saltwater than in freshwater. This is a major source of the toxicity of BKE at a mill pulping a mixture of pine and spruce and contains a number of diterpene alcohols and aldehydes. Elsewhere pimarol, isopimarol, juvabione, and juvabiol were minor toxicants in mechanical pulping wastes (43). The LC50 values of the first two compounds to rainbow trout were both 0.3 mg/l. These diterpene alcohols, together with abienol, 13-epimanool, and (12E)-abienol, constituted the toxic neutral fraction of woodroom effluents from the wet-debarking of spruce, pine, and fir species (45). The toxic diterpene alcohols and aldehydes identified in these various pulping wastes are significant compounds of crude tall oil from Finland (46), Canada (47), and the southern United States (48).

BIODEGRADATION OF DITERPENES

Secondary treatment of BKE is widely practiced in North America, especially where fish populations of commercial or recreational significance inhabit receiving waters. Although aerated stabilization basins are designed to reduce BOD of wastes, they also reduce toxicity (41,49,50). The ability to destroy resin acids in BKE was studied in a bench-scale aerated stabilization basin, and an average reduction of 94% was recorded after three days' treatment (44). The gross content of toxic neutral diterpenes was also decreased (51).

Researchers in France have studied the bacterial degradation of dehydroabietic acid by the microorganism *Flavobacterium resinovorum* (52). This organism was successfully cultivated on dehydroabietic acid as sole carbon and energy source. Intermediate products arose from oxidation at C-3, followed by oxidation at C-7, C-11, and C-12, prior to ring opening. In contrast, degradation of dehydroabietic acid by an unidentified Pseudomonas strain and by *Alcaligenes eutrophus* (53) involved attack at C-7 followed by hydroxylation of the aromatic ring before cleavage of ring C. The degradation products isolated following the action of the pseudomonad or the alcaligene

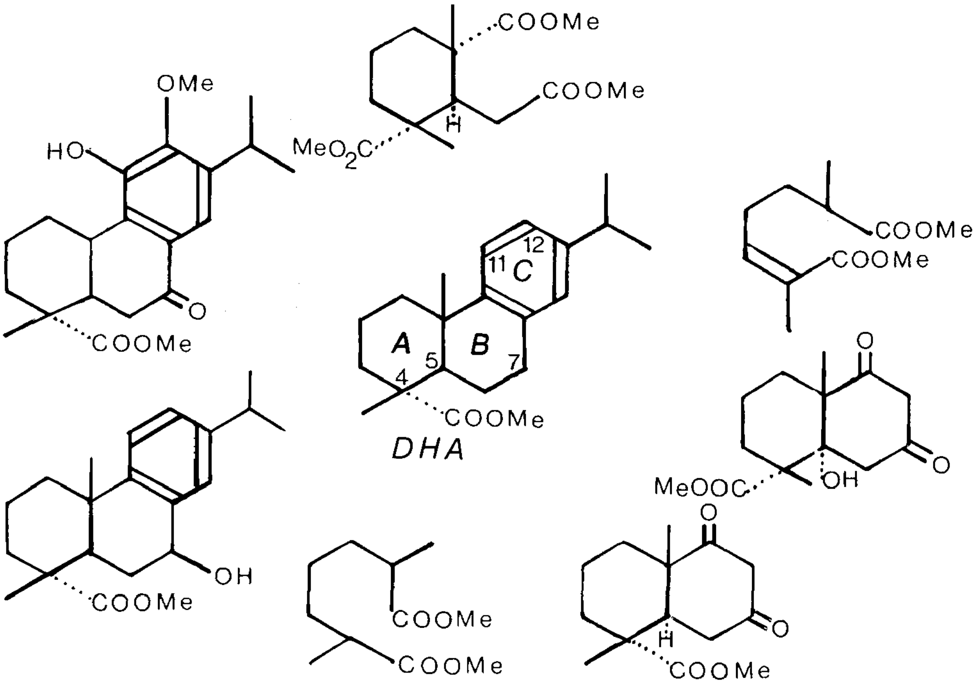

FIG. 1. Metabolites from the action of a Pseudomonad and an Alcaligene on dehydroabietic acid (DHA) as sole carbon source [after Biellmann et al. (53)].

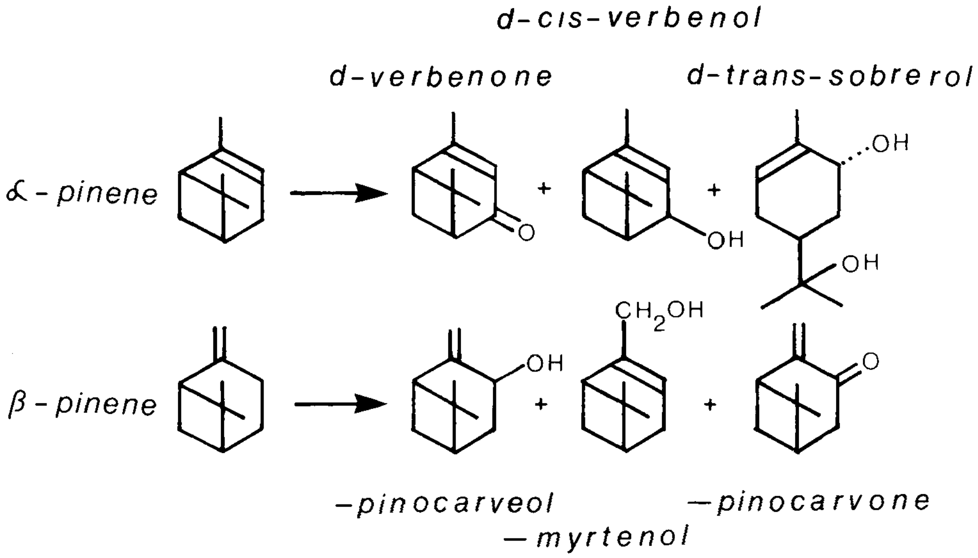

FIG. 2. Metabolites from the action of *Aspergillus niger* in shaken-flask cultures on α- and β-piene [after Bhattacharyya et al. (56,57)].

are shown in Figure 1. These microorganisms were isolated by aerobic enrichment at 28 C in a medium containing dehydroabietic acid. Such organisms are unlikely to play a significant role in receiving waters but might be useful in high-rate activated sludge systems.

The ability of certain yeast and fungal strains to metabolize resin acids from culture media and kraft mill effluents was studied by Spencer et al. (19). Up to 39% of the resin acid content was destroyed by the organisms *Epicoccum nigrum, Rhinocladiella mansonii, Rhodotorula minuta,* and *Trichosporon cutaneum.*

BIODEGRADATION OF MONOTERPENES

Wilson (15,54) has studied the chemical fate of turpentine in aerated lagoons at two kraft mills pulping Douglas-fir. The removal of monoterpenes in one system ranged from 85 to 95% with the highest efficiency in the fall. The major terpene entering the aerated lagoon was α-terpineol which was effectively destroyed. Most compounds were degraded except for the ketones camphor and fenchone which increased across the lagoon, presumably from the oxidation of borneol and fenchyl alcohol. Similar observations were made by Keith in a study of the chemical profiles of the waste-waters of two kraft mills in Georgia (55).

The microbiological oxidation of α- and β-pinene by *Aspergillus niger* was studied by Bhattacharyya et al. (56,57). Various concentrations of d-α-pinene were added to 24 hr shaken-flask cultures, and incubation continued for another 23 hr at 27-28 C. Three oxidized products were obtained in a stereospecific manner, d-verbenone, d-*cis*-verbenol, and d-*trans*-sobrerol (Fig. 2). The optical antipodes of these products resultsed from the incubation of l-α pinene with the same organism. β-Pinene was oxidized to (-)-pinocarveol, (-)-pinocarvone, and (-)-myrtenol (Fig. 2).

RECEIVING WATER PROBLEMS RELATED TO KRAFT DISCHARGES

The most obvious effect of a kraft outfall is the dark brown color imparted to the water column. Current research indicates this is largely an aesthetic problem, but since the color derives mainly from lignin residues, the topic is not relevant here.

FOAM

Unsightly patches of brown foam near outfalls is an environmental problem that may be due to resin acid soaps. Such a foam problem arose on the Thompson River in British Columbia when a large kraft mill at Kamloops was coming on line. This river is famous for its steelhead trout (sea-run strain of rainbows, *Salmo gairdneri*), and the anglers complained of reduced water quality at locations up to 70 miles downstream. A task force investigated changes taking place in the Thompson River and Kamloops Lake (58). Foam samples collected and analyzed during the study period did not contain resin acids or other identifiable compounds characteristic of the pulp mill discharge, and no complaints about foam have arisen recently. This leaves open the question of whether resin acids were released earlier.

Problems have arisen from marine discharges of kraft wastes. In areas of shellfish production, foam containing toxic constituents can drift into intertidal areas with undesirable results (59). Courtright and Bond (60) studied the toxicity of foam to the fluffy sculpin *(Oligocottus snyderi)* and the embryo of the bay mussel *(Mytilus edulis)* in tide pools on the Oregon coast. Foam was four to five times more toxic to mussel larvae than the kraft waste before discharge. Sculpins were least tolerant to KME in low salinity water and most tolerant in seawater of near-normal salinity. It was concluded that foam can adversely affect survival. Although no chemical analyses were performed in the Oregon study to look for resin acids, another investigator has found them in foam from Perdido Bay, Florida, up to 24 miles from the outfall (61). Servizi et al. (62) analyzed highly toxic foam on aerated stabilization basins at two kraft mills. In one case, resin acids and related neutral diterpenes accounted for all the toxicity, whereas in the other 85% was due to a nonionic surfactant pitch control agent of the alkylphenol ethoxylate class.

FISH TAINTING

Kraft effluent discharges can cause tainting problems in resident fish stocks. Shumway et al. (63,64), using a taste panel, studied flavor impairment in salmon or trout exposed to KME. Poor taste and odor scores were recorded after exposure to 1-7% concentrations. Biologically treated effluent caused slight to moderate off-flavor at 16-50%. Similar studies were made on fish in the St. Lawrence River below a kraft mill at Cornwall, Ontario. Evaporator condensate and digester foul condensate were mainly responsible (65). These streams contained volatile organosulfur compounds and turpentine. The problem was ameliorated by steam stripping of condensates and additional weak black liquor oxidation. However, the chemical agents causing tainting were not established by analysis.

In British Columbia, anglers on the Kootenay River downstream of a kraft mill at Skookumchuk have complained of poor flavor of Dolly Varden trout *(Salvelinus malma)* and Rocky Mountain whitefish *(Prosopium williamsoni)* (66). Fishermen on Kamloops Lake and the Thompson River, below Kamloops, have also taken tainted gamefish (58). Fish exposed in cages in the river below the pulp mill outfall and the municipal sewage outfall of the city of Kamloops became tainted. The native peoples at Kitimat say they can no longer utilize eulachons *(Thaleichthys pacificus)* – an oily fish which spawns in the lower reaches of the Kitimat and other coastal rivers. Here biologically treated KME is discharged to the river about five miles from the sea. In none of these examples has the tainting agents been identified, but they are most probably either organosulfur compounds or monoterpenes.

TURPENTINE ESCAPEMENT

Transformation of sulfate turpentine in natural waters was studied at Lake Baikal in the Soviet Union (67). Oxidation took place under conditions of full illumination, and the products included aldehydes, ketones, alcohols, esters, resinous substances, phenols, carboxylic and amino acids. This caused the discoloration of lake water and increased its content of organic substances, nitrates, nitrites, total bacteria, and saprophytic bacteria.

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