

# ENVIRONMENTAL EFFECTS OF APPLYING LIGNOSULFONATE TO ROADS



By

**JAMES W ADAMS**

**DAISHOWA CHEMICALS INC**  
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## SUMMARY:

The overall impact on the environment from applying lignosulfonates to roads is negligible. They are safer to use for stabilisation and dust control than any competing class of chemicals.

The manufacture of lignin involves evaporation; the evaporation process drives off any volatile contaminants such as acetic acid. Corrosion and toxicity towards plants can be readily minimised by pH control and lignosulfonates are non-toxic to animals.

Lignosulfonates have been applied to roads, used in animal feeds and converted into human foods for more than 30 years without problem. This comes as no surprise after learning that the products have the following properties:

- No dioxins present. No other organics present at hazardous levels
- Toxic trace minerals are below EP Toxicity limits \*
- Low order of toxicity towards fish
- Non-toxic orally and non-irritating to the skin or eyes of animals
- No human health problems attributed to exposure
- Very low toxicity towards plants
- Residuals are resistant to decay

When spread on land, there is no risk of contaminating ground water. Published data indicate that at less than 10 kgs per square meter, no problems arise. This is much above the 1 kgm/square meter required for stabilisation and 0.3 kgm used for dust control.

The non-hazardous features and abundant supply of lignosulfonate make it a good material for treating roads (1). It is widely used in Sweden and California – two of the world's most environment-conscious areas.

## PRODUCTION:

In the sulfite pulping of wood, lignin – nature's binder – becomes soluble and is separated from the pulp fibers as spent liquor (SSL). This liquor also contains the naturally occurring wood sugars.

Dilute SSL at 12% solids is treated and concentrated to 50 – 60% solids by evaporating water. Starting from this base material, a family of chemicals has been developed. They are known as lignins or lignosulfonates. They are used in the following applications:

Water Treatment Chemicals	Oil Well Drilling Mud Additives
Components of Adhesives	Textile Dye Dispersants
Animal Feed Pellet Binders	Gypsum Dispersants for Wallboard
Concrete Additives	Leather Tanning Agents
Road Binders	Dispersants for Brick Clay
Feed Molasses Additives	Battery Plate Expanders

*\* As set by the U.S. Environmental Protection Agency*

Of the 1.32 million tons of lignosulfonate sold worldwide, 150,000 tons are sold for application to roadways – 80,000 tons in North America and 70,000 tons in Europe and Australia. The product used on roads is the base product that has not been chemically modified and may be the calcium,

sodium or ammonium form depending on the pulp mill (36). Complex lignosulfonates are sophisticated, high technological content chemicals, showing all the chemical complexity of the naturally occurring lignin from which they were derived.

### **SUGARS:**

The types of sugars present in lignosulfonate will change depending on the kind of wood pulped. Wood from needle-bearing trees yield predominantly the hexoses-mannose, glucose and galactose. Wood from leaf-bearing trees yield predominantly the pentoses-xylose and arabinose. We do not pulp maples!

### **ENVIRONMENTAL EFFECTS:**

#### (a) Dioxins:

During the evaporation of spent liquor, SO<sub>2</sub> and any volatile components are removed. However, with attention recently focused (38) on the presence of dioxins in paper mill effluents, the U.S. Environmental Protection Agency approved laboratory – Enseco Inc., was contracted to determine the dioxin content of SSL roadbinder. Results from analysing a 7-day composite of Lignosol B, (a calcium lignosulfonate) (37) show that it contains no detectable amounts of 2, 3, 7, 8 – tetrachloro dibenzofuran or 2,3 7, 8 – tetrachloro dibenzop-dioxin. This is not surprising as chlorinated organic contaminants require the presence of substantial chlorine concentrations. This can come from:

- Using chlorinated biocides after bleaching
- Bleaching fibers with chlorine, hypochlorite or chlorine dioxide

But the Quebec mill (REED) does not bleach. Mills that do bleach only do so after the lignosulfonate has been separated and removed from the system.

#### (b) Trace Elements:

Lignosulfonates are wood extracts that contain the mineral elements naturally present in trees. Depending on the tree type and soil on which it was grown, the wood contains different kinds and amounts of minerals. Element analysis of tree barks that are typical for plant residues were published by Harder and Einspahr (12).

The U.S. Environmental Protection Agency (EPA) has recently defined the maximum concentration of elements to establish EP Toxicity (5). Limits are shown in Table 1.

**TABLE I.****MAXIMUM CONCENTRATION OF CONTAMINANTS FOR CHARACTERISTIC OF EP TOXICITY**

EPA Hazardous Waste No.	Contaminant	Maximum Concentration, ppm
D004	Arsenic	<b>5.0</b>
D005	Barium	<b>100.0</b>
D006	Cadmium	<b>1.0</b>
D007	Chromium	<b>5.0</b>
D008	Lead	<b>5.0</b>
D009	Mercury	<b>0.2</b>
D010	Selenium	<b>1.0</b>
D011	Silver	<b>5.0</b>

REED's lignosulfonate products are submitted periodically for Proton Induced X-ray Emission analyses. The testing procedures can detect all eight of the EP Toxicity contaminants plus 67 others. Results on REED products (7) are reported in Table II. Note that barium, cadmium, mercury, selenium and silver could not be detected. The other EP Toxic elements are present at levels below the maximum concentration.

**TABLE II.****TRACE ELEMENTS IN LIGNOSULFONATES****Reed Calcium Lignosulfonate**

	Concentrate	Neutralized Concentrate
	ppm in solids	
Titanium	5.6	-
Aluminum	-	-
Silicon	105	241
Phosphorus	540	516
Manganese	118	155
Iron	207	869
Cobalt	1.8	6.6
Nickel	0.9	1.4
Copper	2	1.2
Zinc	25	31
Gallium	0.3	-
Bromine	0.7	0
Rubidium	3.7	2.5
Strontium	32.5	34
Zirconium	1.8	1.6
Molybdenum	0	2.5
Arsenic	0.1	-
Barium	-	-
Cadmium	-	-
Chromium	-	2.7
Lead	-	0.3
Mercury	-	-
Selenium	-	-
Silver	-	-

Neutralisation adds some trace minerals as they are contributed by the naturally occurring limestone that is used in the neutralising process.

### ACUTE ANIMAL TOXICITY:

During the past 22 years, many of REED LIGNIN'S products have been examined. All of them proved to be non-toxic. Table III shows the results for 18 widely used products. The basic lignin used on roads would be even more innocuous (22).

To assess health risks involved in handling a new chemical, it is tested for acute toxicity in white rats and skin and eye irritation on rabbits. If all 10 rats in a test survive a one-time feeding dose of five grams/kilogram of body weight, the product is considered non-toxic according to the Occupational Safety and Health Administration (OSHA) rule, printed in the U.S. Code of Federal Regulations 29 CFR 1910.1200 (4). If rats do not survive the five gram/kilogram feeding level, testing is conducted to find the dose that kills 50% of the rats (LD<sub>50</sub>).

Empirical scores are used to report results for skin and eye irritation tests with a skin irritation score of less than five needed to rate a material non-irritating to the skin. Testing for eye irritation is done following instructions in the Code of Federal Regulations 16 CFR 1500.42. The Hazelton Laboratory report on testing the lignin product, Additive-A Type 3, (10), is typical for doing this kind of work.

**TABLE III**  
**ACUTE TOXICITY OF LIGNIN PRODUCTS**  
**TYPICAL REED PRODUCTS**

PRODUCT	TEST LAB	DATE	ORAL TOXICITY ON WHITE RATS, LD <sub>50</sub> g/kg	RABBIT SKIN IRRITATION INDEX	RABBIT EYE IRRITATION SCORE		
					24 hrs	48 hrs	72 hrs
Maracell XE	WARF	1977	> 5	0	0.33	0	0
Marasperse N-22	WARF	1975		0	0	0	0
Marasperse CB	WARF	1975		0	0	0	0
Marasperse CE-22	WARF	1977	> 5	0.17	2.0	0	0
Marasperse B-22	WARF	1975		0	0	0	0
Marasperse OS	Raltech	1978	> 5	0	0	0	0
Marasperse N-3	Raltech	1978	> 5	0	0	0	0
Marasperse CBOs-3	WARF	1977	> 5				
Marasperse CR-23-7	WARF	1977	> 5	0.21	0.67	0	0
Marasperse XCB-2	WARF	1977		0.17	0.67	0	0
Norlig 41	WARF	1975		0	0	0	0
Norlig 41d	WARF	1975		0	0	0	0
Norlig 41N	WARF	1976	> 5	0	0	0	0
Norlig 412	Raltech	1978	> 5	0.2	0	0	0
Additive-A, Type 3	Hazelton	1985	> 5	1.5	0	0	0
Americo SD-1	Raltech	1978	> 5	0	0	0	0
Kelig 32	WARF	1977		0	0	0	0
Kelig 32-2	Raltech	1978	> 5	0.1	0	0	0

Reed's roadbinder is among the Norlig 41 class of products.

### SUBACUTE TOXICITY:

In 1960, American Cyanamid, Hercules Powder, NOPCO Chemical and Socony Mobil Oil Companies sponsored a study at the Industrial Bio-Test Laboratories to Determine the 90-day subacute oral toxicity of a REED sodium lignosulfonate, Marasperse N.

Seven groups of 14 white rats were fed chow containing 0, 0, .01, .05, .20, 1.0 and 5.0% Marasperse N for 90 days. Results were reported by Kay and Calandra (15).

- None of the five dietary levels of Marasperse N had adverse effects on the growth of rats
- Only small differences were found for food consumption and food utilisation
- No dose-correlated trends were apparent in mortality data
- No significant abnormal behaviour was noted among animals of any group
- Periodic blood studies disclosed no adverse findings
- Urine analysis revealed no significant abnormalities
- No significant gross pathological findings were noted at autopsy of sacrificed animals

### **CHRONIC TOXICITY:**

For effects of long-term exposure there are the good health records for those who have worked with reacting and spray drying this material. Over a period of 40 years to date, no human health problem has been attributed to exposure to lignosulfonate.

Before using lignosulfonate in poultry and animal feeds, calcium, sodium and ammonium forms of SSL were scrutinised very carefully for long-term health effects. Results of studies conducted to get approval to use the SSL products in animal feeds were obtained from the U.S. Department of Health & Human Services under the Freedom of Information Act (25). After reviewing data filed with Food Additive Petitions No. 706 and 1239, the Commissioner of the Food & Drug Administration ruled on August 15, 1962 that:

“.....The Division of Food concludes that the product is reproducible, of uniform composition, and that it accomplishes its intended technical effect of pellet binding. An analytical method is available for regulatory purposes.

The Division of Pharmacology concludes that use of the additive in animal feed under the proposed conditions of use in animal feed is safe.

The Division of Veterinary Medicine concludes that the additive is safe for use in animal feeds and effective as a pelleting aid.

Mr Philbeck, Meat Inspection Division, ARS, U.S.D.A., offers no adverse comment to this order.”

Today, 30 years after lignins were introduced into animal feeds, no chronic toxicity problems have arisen. This is not surprising, after all – hay, grain and forage contain 20% lignin.

## FISH TOXICITY:

Because a lot of water is needed for transporting, processing and pulping wood, paper mills locate near oceans, lakes and rivers. Many studies have been conducted to measure the water polluting effects of the following:

- Wood transport
- Debarking
- Pulping
- Fiber washing
- Fiber bleaching
- Paper making (6, 20, 21, 31, 32)

But, most of the studies have been concerned with spent liquor prior to evaporation, whereas lignosulfonate products, sold for road dedusting are SSL concentrates. When removing water in evaporators sulfur dioxide, acetic and formic acids are distilled off leaving liquors that are less toxic than the raw material. Published information about various lignosulfonates are summarised in Table IV along with reference toxicity values for familiar products.

Evaporating water and volatiles from dilute calcium and sodium SSL substantially reduces toxicity to the point where fish can tolerate 3,000 rather than 1,000 mg/l of solids. With purified lignosulfonates the tolerance is increased to 7,000 mg/l (27). The common salts calcium chloride, sodium chloride and sodium sulfate have LC<sub>50</sub> 96-hour values in the 5,000 – 10,000 range (8). In reference books (18,28) the cutoff point for LC<sub>50</sub> 96-hour values is 1,000 mg/l. Compounds with values greater than this are given a value of >1,000 and are considered to have a low order of toxicity. Calcium and sodium lignosulfonates, calcium chloride, sodium chloride and sodium sulfate all are in this category. By contrast, the popular laundry detergent **Tide** and surfactants used in making such products are very toxic toward fish (14). Values ranging from 4 – 50mg/l are reported for these materials in Table IV.

**TABLE IV**

### **TOXICITY OF WATER SOLUBLE MATERIALS TOWARD FISH**

REFERENCE	FISH	LIGNOSULFONATE TESTED	LC <sub>50</sub> – 96 hours mg solids/litre
Wilson	Rainbow Trout	Lignosol BD (Quebec)	3,700
Stapanian	Rainbow Trout	Lignosite (West Coast)	2,125
Wilson	Rainbow Trout	Lignosol XD	3,500
Roald	Rainbow Trout	Purified Na Lignosulfonate	7,300
Canada	Various	Calcium Chloride	5,000
Jones	Various	Sodium Chloride	6,000
Jones	Fathead Minnows	Sodium Sulfate	9,000
Jones	Fathead Minnows	<b>Tide</b> Laundry Detergent	50
Jones	Fathead Minnows	Sodium Dodecylbenzene Sulfonate	4
Jones	Fathead Minnows	Sodium Lauryl Sulfate	5

## PLANTS:

With any liquid material that is applied to roadways, there is concern about harming plants that grow near the roads. Treating dirt roads with lignosulfonate to stabilise them for carrying vehicle traffic requires mixing 15 tons of solid per mile in the top six inches of soil on a 24-foot wide road. To do this, lignosulfonates are diluted with water. After spraying on the roadway, a road grader or scarifier is used to mix the binder with soil. Compacting with a roller is done soon after mixing.

For dust control, only 2.5 tons of solids per mile are sprayed on premoistened dirt roads. This total amount is usually applied in two applications.

In recent years both techniques have been used in Europe and North America. They account for the sale of 150,000 short tons of lignosulfonate solids that are used to treat 20,000 miles of roadway. No complaint of plant damage has been reported from this wide exposure (30). This is not a surprise after reading details of a very bold experiment that Stapanian and Shea (29) conducted. They applied Lignosite, a 50% solids calcium lignosulfonate, directly to the ground cover of Douglas Fir plantations in the state of Washington. Very high application rates of 21, 42 and 63 tons of solids per acre were applied to twelve 5 x 10m (16 x 33 feet) plots of forest land. Four plots received the same treatment. These treating levels are way above the 5 and 1.3 tons of solids per acre applications used for stabilising and dedusting roads.

Observations made periodically up to 12 weeks after the Lignosite was applied, indicated that the woody vegetation was not affected. The biomass of herbaceous plants was significantly decreased only at the two highest application rates. The growth of Douglas Fir trees was not significantly affected.

In another study Gast and Early (9) reported on the phytotoxicity of three commercial lignosulfonates toward garden crops, cotton and tobacco. This work was done with ingredients used for pesticide formulations. Of the 77 products tested, Marasperse C, Marasperse CB and Marasperse N were the least toxic. Pertinent results are reproduced in Table V. Lignosulfonates are now widely used in herbicide and pesticide formulations.

Chlorides are not expensive in California and, like lignins, are often used for dust abatement. However, at relatively low dosage chlorides can kill vines and care is suggested when applying them (19).



**TABLE V**  
**PHYTOTOXICITY RATINGS**  
**Plant Injury**

**0 = None, 1 = Slight, 2 = Moderate, 3 = Heavy, 4 = Severe**

Plant	Conc in Water Applied %	Marasperse C (Calcium Lignosulfonate)	Marasperse CB (Sodium Lignosulfonate)	Marasperse N (Sodium Lignosulfonate)
<b>LEAVES</b>				
Bean	1.0	0	1	0
	0.1	0	0	0
Corn	1.0	0	0	0
	0.1	0	0	0
Cotton	1.0	0	0	0
	0.1	0	0	0
Cucumber	1.0	0	0	0
	0.1	0	0	0
Tobacco	1.0	0	0	0
	0.1	0	0	0
Tomato	1.0	0.5	0.3	0
	0.1	0	0	0
<b>ROOTS</b>				
Tomato	1.0	4	1	4
	0.1	0	0	0
	0.01	0	0	0
	0.001	0	0	0
Tobacco	1.0	0	0	0
	0.1	0	0	0

**GROUNDWATER:**

The effect of lignosulfonate on groundwater is related to concentration. Applying 4,000 tons of dissolved solids each year to the same acre of porous soil will drive the liquor down into the groundwater with no time for destruction by fermentation. Results from ponding studies are vivid demonstrations. With much more modest one-time applications of 20 – 60 tons / acre Stapanian (29) indicates slow movement and time for fermentation which should pose no threat to contaminating groundwater. With no serious groundwater contamination expected at 50 tons/acre, applying 5 and 1.3 tons/acre for road paving and dust control would not be a serious matter.

**DECAY / BIOLOGICAL OXYGEN DEMAND:**

Sugars and carbohydrates are easily fermented by many different microbes. With as much as 35% of these easily fermented substances present in lignosulfonate, and few toxic materials around, it is no surprise that partial decay occurs quickly. In fact, lignosulfonate is used as a fermentation media for the commercial production of ethyl alcohol, fodder yeast and food-grade yeast.

Because of the wood sugars, lignosulfonates are not added to waterways containing fish and a marginal supply of dissolved oxygen. Microbes will feed on the sugars and consume oxygen in the process.

Results of many tests on basic lignosulfonate indicate that its five-day Biochemical Oxygen Demand (BOD<sub>5</sub>) is 0.23 pounds per pound of solids. Microbes acting on 100 pounds of basic lignosulfonate for five days require 23 pounds of oxygen. The 100 pounds of lignin must be mixed with 394,000 gallons (1,500 cubic meters) of water containing 11 ppm dissolved oxygen to wind up after five days with water containing 4 ppm dissolved oxygen. The 4 ppm dissolved oxygen is where fish kill first appears. This means that 30 ppm of SSL solids will allow dissolved oxygen at a safe level. More complex lignosulfonates show lower BOD<sub>5</sub> values.

During the 1960s, much attention was given to laundry detergents that produce voluminous and persistent foam in discharge waterways. The problem was traced to sudsing surfactants that biodegraded very slowly. In developing new sudsing surfactants, River Dieaway was the most popular test used for measuring biodegradation. The test was applied to examine lignosulfonates.

Small amounts of lignosulfonate, dissolved in aerated Wisconsin River water, were held at room temperature for 33 days. Samples removed periodically were tested for organic matter content by oxidising with dichromate to determine the Chemical Oxygen Demand (COD). Data showed that Norlig A, a lignosulfonate roadbinding material, degraded 28% in five days and 43% in 33 days. This closely corresponds to the carbohydrate content of the product, and indicates that pure lignin resists decay. Pure lignin is the last to go in natural decay processes.

The 54% material remaining after 33 days of incubation persists as a roadbinder, and will show as natural lignin colour in waterways. It is a situation much like the brown colour imparted to streams and lakes by humus and humic acids produced in the decay of plants and trees.

### **CORROSION:**

Severe corrosion of automobile underparts is usually associated with the use of salts for de-icing roads during winter months. This is not always so. A study by the Swedish Highway Department (17), indicates that in areas where calcium chloride or magnesium chloride is used for keeping dust down on dirt roads, automobile corrosion from this source is more severe than from salts used for road deicing(16).

Because of the nature of their binding mechanism, lignins absorb onto soils and clays and form cohesive bonds. This is very different from calcium chloride which is loosely concentrated on the surface of the road from where it is easily blown into passing vehicles where it can start its insidious corroding activity.

### **DISCUSSION:**

This survey of lignin and the environment has only touched on the subject. However, a full reference index is attached for those wishing to study the subject in greater depth.

Any open minded analysis of the data will conclude that the use of lignin for dust control and road stabilisation is not only environmentally safe, but is more “environmentally friendly” than any other dust palliatives available.

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