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Treatment of drinking water to remove color, turbidity, and other impurities is increasing steadily. Water treatment plants in Connecticut produce about 80,000 tons of alum sludge as a by-product annually. The sludge contains about 5 percent solids, and Connecticut regulations prohibit disposal of any liquid waste containing less than 20 percent solids in landfills. Therefore, alum sludge presents a severe disposal problem for water utilities.

Alum sludge does not give up water readily, so it is often transported in tank trucks for storage in lagoons or spread on land to dry. After freezing, the sludge dries more readily, and can then be transported to landfills for disposal.

Because of the amount produced and the difficulties and expense in handling and disposal, we investigated possible uses of alum sludge as a soil amendment.

Properties of Sludge

Recent concern over the possible effects of acid rain on mobilization of aluminum in soil and water suggests that a brief review of aluminum chemistry would be helpful in understanding the reactions of alum sludge that is disposed.

Aluminum is the most abundant metallic element in the earth's crust. Moreover, hydrolysis of the aluminum ion during natural weathering and soil formation produces a moderately acidic environment. Thus, many of the properties of acid soils in humid temperate climates such as in Connecticut are controlled by the chemistry of aluminum. As a consequence, soil scientists have studied the chemistry of aluminum in both soil and water in considerable detail and a number of review articles are available (Coleman and Thomas, 1967; Frink, 1973).

The term alum sludge is a slight misnomer. Technically, alum is a mixed aluminum sulfate salt with the general composition M $Al(SO_4)_2\ 12H_2O$ where M is either the potassium or ammonium ion. In water treatment plants, commercial grade liquid aluminum sulfate $Al_2(SO_4)_3 \times H_2O$ is generally referred to as alum. Alum clarifies water by hydrolysis of aluminum to form a gelatinous precipitate of aluminum hydroxide. Although alum sludge is an aluminum hydroxide sludge, the common usage will be followed here.

When an aluminum salt is added to water, as in a water treatment plant, the aluminum ion (Al^{+3}) hydrolyses with water to produce acid (H^+) as shown below:

$$Al^{+3} + 3H_2O = Al(OH)_3 + 3H^+.$$

Thus, aluminum sulfate ($Al_2(SO_4)_3$) added to water produces aluminum hydroxide ($Al(OH)_3$) and the resulting hydrogen ions (H^+) produce an equivalent amount of sulfuric acid (H_2SO_4). Hence, aluminum salts can be considered acids, and it is impossible to distinguish between the two sources of acidity in any system containing aluminum and added acid. If water contains sufficient neutralizing capacity in the form of dissolved bicarbonate or other sources of alkalinity (OH^-), the acidity is neutralized and a gelatinous precipitate, aluminum hydroxide ($Al(OH)_3$), which is known as alum sludge, forms.

This same reaction occurs when limestone is added to soils to counteract natural soil acidification. It can be described by rewriting the previous equation (omitting the hydrolysis step for simplification):

$$Al^{+3} + 3OH^- = Al(OH)_3$$

Conversely, $Al(OH)_3$ can react with acid according to:

$$Al(OH)_3 + 3H^+ = Al^{+3} + 3H_2O$$

This is the basis for using aluminum hydroxide in antacids. These two reactions show how aluminum can buffer soil and water against changes in acidity.

The ultimate product of the neutralization of aluminum is a crystalline form of $Al(OH)_3$ known as gibbsite. The presence of gibbsite is important in controlling the concentrations of aluminum in soil and water since, at any given pH, the concentration of aluminum can be predicted from a knowledge of the solubility of gibbsite (Frink and Peech, 1962).

In soils, equilibrium with gibbsite is not always observed since there are several intermediate forms of aluminum hydroxide that are less highly crystalline and hence somewhat more soluble. Because there are also some polymeric forms of aluminum that are not completely neutralized, the resulting precipitate of aluminum hydroxide may carry a small net positive charge. This residual positive charge greatly affects the reactions of phosphorus with soil and water. Because phosphorus is normally present in the form of negatively charged anions, it is readily sorbed by the positively charged aluminum hydroxide. This reaction then controls the availability of phosphorus for plant growth in acid soils. It also is the principal mechanism for removal of phosphorus from waste water in sewage treatment plants and is the basis for removal of phosphorus from eutrophic lakes by treatment with alum (Norvell, 1982).

Analyses of Sludge

The results of EPA elutriate tests on sludge from the Saltonstall plant and the West River plant follow:

Parameter	Saltonstall	West River
	mg/l	
Arsenic	< 0.01	< 0.01
Barium	0.21	0.1
Cadmium	< 0.005	< 0.005
Chromium (Hex)	< 0.01	< 0.01
Lead	< 0.01	< 0.01
Mercury	< 0.001	< 0.001
Selenium	0.09	< 0.01
Silver	< 0.01	0.01
	ppb	
Endrin	< 0.2	< 0.2
Lindane	< 4	< 4
Methoxychlor	< 100	< 100
Toxaphene	< 5	< 5
2,4-D	< 100	< 100
2,4,5,-T (Silvex)	< 10	< 10
	%	
Total Solids	11.3	9.7
Volatile Solids	24.0	33.4
Fixed Solids	76.0	66.6

Samples extracted for 24 hrs at pH 5.0 and tested according to EPA methods. Source: South Central Connecticut Regional Water Authority.

Additional elutriate tests were performed on dried Saltonstall sludge (SM) and soil adjacent to a disposal site (CM), as well as on sludge from West River (SB) and adjacent soil (CB), and the results follow:

Parameter	SM	CM	SB	CB
		mg/l		
Copper	< .02	< .02	.17	<.02
Zinc	.05	.16	.04	.06
Iron	< .02	2.80	.34	3.28
Manganese	1.97	.97	16.47	1.54
Aluminum	< .2	.68	.91	1.14
Nitrate N	.08	.35	.05	.05
Nitrite N	.003	.002	.003	.004
Total Phosphorus	.013	.013	.013	.014

Samples extracted for 24 hrs at pH 5.0 and tested according to EPA methods. Source: South Central Connecticut Regional Water Authority.

The concentrations of aluminum in the extract at pH 5 allows calculation of the apparent solubility of $Al(OH)_3$ in these samples. The results show that all four samples contain a form of aluminum that is slightly more soluble than gibbsite. Since these results are typical of the concentrations of aluminum found in soils at pH 5 (Frink and Peech, 1962) they do not suggest aluminum or other metal toxicity in either the sludge (S) or the soil (C).

As for the possible effects of acid rain on alum sludge, calculations show that the amount of strong acid in a year of rainfall in Connecticut at pH 4.0 can be neutralized by about 50 lb of limestone per acre (Krug and Frink, 1983). Similar calculations for alum sludge show that a year of acid rain could dissolve (and hence be neutralized by) about 25 lb of alum sludge per acre. Thus, mobilization of aluminum by acid rain landing on alum sludge would be negligible.

The chemical analyses reveal that although the alum sludge contains few if any plant nutrients, it contains 25-35% organic matter (volatile solids as measured by loss on ignition). Some organic polymer is added to the alum to speed flocculation. Sludge from the West River plant contains a higher ratio of polymer to alum than sludge from the Saltonstall plant as indicated by the larger amount of volatile solids in the West River sludge. Samples of dried and aged sludge analyzed by X-ray diffraction showed only the diffuse diffraction patterns typical of amorphous aluminum hydroxide.

Disposal of Sludge

Many processes for dewatering sludge have been examined (Westerhoff and Daly, 1974). However, all are costly and, with the exception of alum recovery, they require disposal of the dewatered sludge in a landfill. Westerhoff and Daly (1974) also expressed concern that runoff from sludge in a sanitary landfill could contain soluble aluminum.

Although land application of lime sludge has been suggested (Russell, 1975), similar use of alum sludge from water treatment plants has not been reported. In tertiary sewage treatment plants, calcium hydroxide, aluminum sulfate or ferric chloride is used to remove phosphorus. Soon, Bates and Moyer (1978, 1980) and Soon *et al.*, (1978) studied the effects on plants and soils of sewage sludge treated with these three chemicals. Treatment with calcium hydroxide tended to increase soil pH, and treatment with iron tended to decrease soil pH, although this was confounded with the acidifying effects of nitrification. Soon, Bates and Moyer (1978) concluded that with acid soils, treatment with calcium would be best. Gestring and Jarrell (1982) treated a sludge with aluminum sulfate, ferric chloride or calcium hydroxide in amounts required to provide coagulation. They measured the uptake of phosphorus and the heavy metals zinc, manganese and cadmium by Swiss chard *(Beta vulgaris)*. The only effect noted was at the highest rates of application. In this case, the aluminum treated sludge acidified the soil and increased uptake of the metals. Hence, they warned against using such sludges on acid soils.

Alum sludge is generally produced by adjustment of pH of the water to provide optimum flocculation. Drying,

Table 1. Physical properties and plant growth in potting media amended with alum sludge.

Treatments					Physical Properties							
	%/V.				%/V.				g/cc	g/plant	mg/l*	%
#	Soil	perlite	peat	alum	air at c.c.	avail. H_2O	Unavail. H_2O	total pore s.	bulk density	lettuce growth	soil P	tissue P
1	33	33	33	0	7.7	45.6	16.9	70.2	0.52	5.9	95	.52
2	33	33	0	33	8.9	34.5	22.4	65.8	0.63	4.6	42	.14
3	33	0	33	33	6.6	38.7	24.1	69.4	0.60	4.5	39	.08
4	0	33	33	33	16.8	50.2	14.4	81.4	0.27	5.8	26	.09
5	25	25	25	25	6.8	45.3	20.0	72.1	0.54	3.5	34	.11
6	17	17	17	50	7.2	45.8	22.6	75.6	0.48	2.9	30	.08
7	11	11	11	67	13.9	40.3	23.7	77.9	0.43	2.0	33	.09

*** mean of six tests run throughout growth period**

particularly following freezing (Wilhelm and Silverblatt, 1976), produces a highly granular material that resists rewetting. Tertiary treatment of sewage effluent with alum generally does not involve adjustment of pH, and the resulting alum sludge is usually mixed with sewage sludge and cannot be distinguished from it. Hence, differences observed when sludges are applied to soil may be attributed to differences in pH and crystal form of the aluminum hydroxide.

Experimental

Greenhouse and Field Experiments

In preliminary experiments in the greenhouse, we germinated ryegrass on mixtures of soil and alum containing 0, 25, 50, 75, and 100% dried alum sludge. As expected, the sludge increased soil pH from 5.6 in the control to 6.2 in the 100% alum treatment. Although the sludge did not hinder seed germination, we noted some inhibition of growth.

Thus, we conducted several experiments with two objectives: the first was to substitute dried alum sludge for various constituents in potting soil mixtures, and to measure their ability to support plant growth. Potting mixtures were prepared and test plants grown and analyzed as in earlier experiments (Bugbee and Frink, 1983)

The second objective was to spray wet alum sludge on forest plots and measure effects on soil, litter decomposition, and tree growth. The plots were established in cooperation with the South Central Connecticut Regional Water Authority in the watershed of Lake Gaillard in North Branford, Connecticut.

Alum as an Amendment for Potting Media

Effects on physical properties. Potting media containing equal portions of soil, perlite and peat moss are often used by growers of greenhouse crops. Recent studies suggest that potting soils could be improved by the addition of materials that increase their aeration and available moisture holding capacity. To determine if dried alum would be a suitable material, media with seven combinations of alum, soil, perlite and peat moss were prepared as shown in Table 1. The alum came from the Lake Saltonstall plant and had dried over the winter. Treatment 1 contained no alum. In treatments 2, 3 and 4 alum replaced either soil, peat or perlite. In treatments 5, 6 and 7 alum was added in different proportions to equal amounts of soil, perlite and peat.

The physical properties of each treatment, based on the mean of five replicates, are shown in Table 1. Media aeration (air at container capacity) was significantly improved in treatment 4, when alum replaced soil, and in treatment 7, when equal portions of soil, perlite and peat were mixed with 67% alum. Aerations of 16.8 and 13.9 percent, respectively, fall within an optimum range of 10-25 percent suggested by previous Station research. The quantity of available water improved where alum replaced soil in treatment 4. Differences in unavailable water, total pore space and bulk density appeared to be minimal among treatments.

Lettuce (*Lactuca sativa*, cv. iceberg) seedlings were transplanted into 4½" standard pots arranged in the greenhouse in a 7 x 7 latin square surrounded by a border row. Conventional amounts of nitrogen, phosphorus potassium and micronutrient fertilizers were added. Pronounced differences in plant growth became apparent after two months. Most noticeably, lettuce growing in media amended with alum took on a purple hue. Purple coloration of leaves is usually associated with inadequate available phosphorus. Soil tests and leaf tissue tests (Table 1) confirmed that phosphorus was less available to plants in media containing alum. Plant growth, as determined by the mean oven dry weight of the plants from each treatment (Table 1), showed a significant decrease in all media amended with alum except treatment 4. The increase in plant growth in treatment 4 can likely be explained by improvements in aeration and moisture holding capacity, which overcame the adverse effects of phosphorus deficiency.

We concluded that dried alum can improve the aeration and available moisture holding capacity of a less than optimum potting media. Deficiencies in plant-available phosphorus that occurred in media amended with alum were probably due to phosphorus fixation by aluminum.

Effects of Phosphorus Fertilization. A second experiment determined the extent of the phosphorus deficiencies. Two methods were used: doubling the initial rate of phosphorus fertilization and adjusting the volume percentage of alum added.

Alum that had dried over the winter was added to a one part peat - one part vermiculite media at rates of 0, 5, 10,

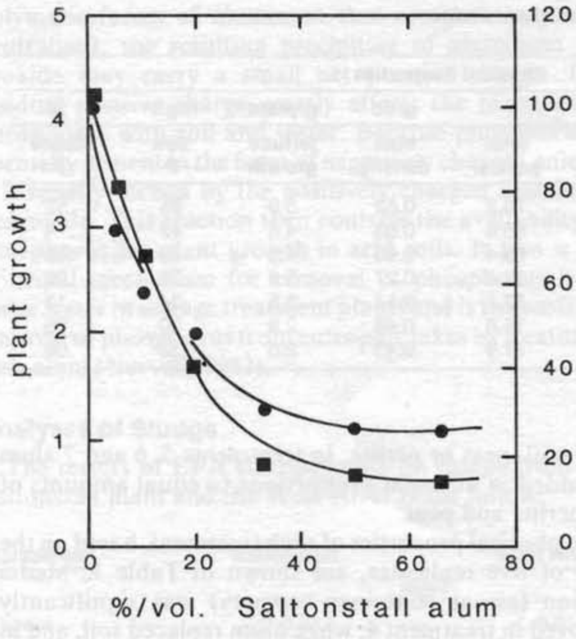

Fig. 1. Growth of marigolds (■) and available soil phosphorus (●) in potting media amended with alum sludge from the Saltonstall Plant.

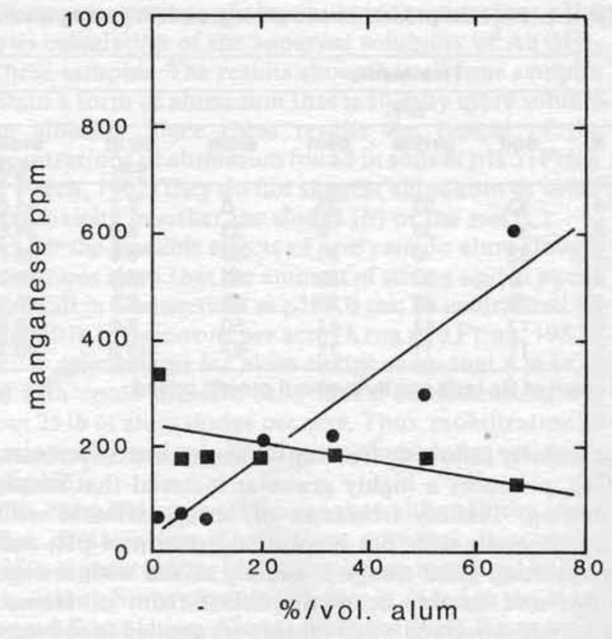

Fig. 3. Concentration of manganese in marigold tissue grown in potting media amended with alum sludge from the Saltonstall Plant (■) and the West River Plant (●).

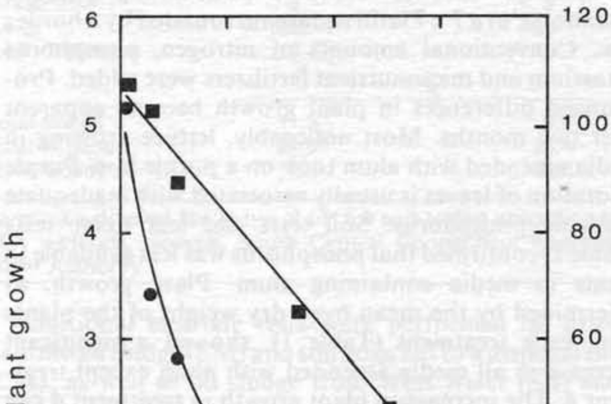

Fig. 2. Growth of marigolds (■) and available soil phosphorus (●) in potting media amended with alum sludge from the West River Plant.

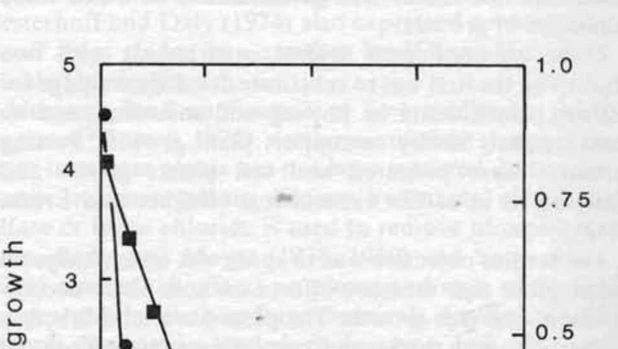

Fig. 4. Concentration of phosphorus (●) in marigold tissue and growth of marigolds (■) in potting media amended with alum sludge from the Saltonstall Plant.

20, 33, 50, and 66 percent by volume. Conventional amounts of nitrogen, potassium and micronutrient fertilizers and twice the normal amounts of phosphorus were added. Marigolds (*Tagetes* cv. lemondrop) were grown using methods similar to those of the previous experiment.

Figures 1 and 2 illustrate how plant growth and available phosphorus responded to various volume percentages of alum. Significant declines in growth occurred at all levels of alum. The only difference between the alum sources was a curvilinear decrease in growth with alum from Saltonstall, and a linear decrease in growth with alum from West River. Available phosphorus in the media, as determined by the Morgan Soil Test, declined curvilinearly as the volume percentage of alum from both sources increased.

Soil tests for nitrate, ammonium, potassium, calcium, and magnesium revealed higher, but not toxic, levels of both elements in media high in alum. Manganese was higher in media containing alum from West River. Analysis of the marigold tissue for nutrients including calcium, magnesium, manganese and potassium suggested that only manganese was affected by increased proportions of alum. Figure 3 illustrates the relationship between manganese in the tissue and alum in the media. Alum from Saltonstall marginally decreased the uptake of manganese while alum from West River significantly increased the uptake of manganese. The likely explanation is that alum from West River contained about eight times more extractable manganese than alum from Saltonstall. Growth, however, did not seem to be associated with differences in manganese. Moreover, the manganese concentrations were similar to those found in tomatoes grown in acid soils where no symptoms of manganese toxicity were observed (Peaslee and Frink, 1969).

Figures 4 and 5 demonstrate the correlation between the decline in plant growth and phosphorus in the plant tissue at the end of the experiment. The relation for Saltonstall alum is shown in Figure 4 and West River alum in Figure 5. The curvilinear decline in tissue phosphorus as alum rates from both sources increased corresponds well to the decline of available phosphorus in the media.

We draw these conclusions from the experiments with potting media:

- Phosphorus deficiencies caused by addition of dried alum cannot likely be overcome by doubling the initial phosphorus fertilization.
- Alum from Saltonstall and West River produced similar declines in plant growth indicating little difference between the two sources.
- No direct effects on plant growth due to manganese were noticed, although uptake of manganese may be affected by the source of alum.

Disposal of Liquid Alum on Forested Land

To test the effects of liquid alum sludge applied to forested land, we established four 15 m x 11 m plots in both a deciduous and coniferous forest. The deciduous forest was predominantly sugar maple (*Acer saggarinum*) and the coniferous forest was a monostand of Eastern hemlock (*Tsuga canadensis*). The average age of the trees was 25 to 50 years.

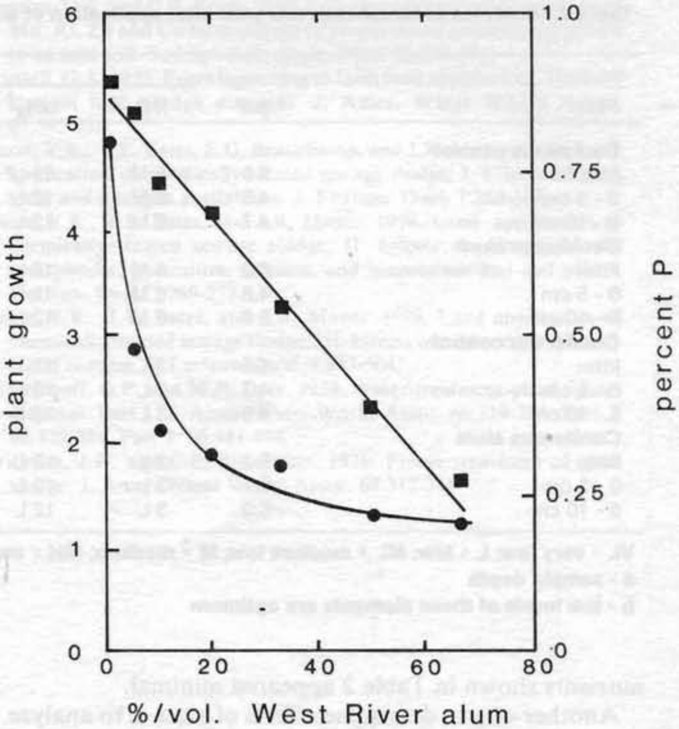

Fig. 5. Concentration of phosphorus (●) in marigold tissue and growth of marigolds (■) in potting media amended with alum sludge from the West River Plant.

Two plots in each forest were treated with the equivalent of 1170 m³/ha (124,800 gal/acre) of liquid alum, which contained approximately 1.5% solids for a total of 17,500 kg/ha of dry alum (15,725 lbs/acre). The wet alum sludge was from the Saltonstall plant. Half the alum was applied in fall 1983 and the remainder in spring 1984. The alum was sprayed from a fire hose connected to a tank truck. A centrifugal pump increased the pressure and decreased the time needed for application. In preliminary tests, the sludge tended to run off and accumulate at the low end of a plot on recently plowed ground with about a 5% slope. This was not a problem in the forest plots, in part because the slope was less (0-3%), but water infiltrated faster in the forest floor than in the bare soil.

Prior to the alum application, soil samples were taken from each plot. In general, the tests revealed the soils were acidic and infertile, a condition typical of most Connecticut forests. Each tree was marked with a number and its diameter at breast height (DBH) measured.

Approximately one year after the initial alum application, the DBH of the trees was remeasured, soil samples were taken, and leaves and needles were analyzed for uptake of nutrients. No tree damage had occurred and little residue of alum was visible. There were no significant changes in DBH between treatments.

In the plots that received alum, however, the soil pH increased. Since the pH of alum is near 7.0 and the forest soil is below 5.0, this agrees with our expectations. In general, alum raised the pH by 0.5 to 1.0 in the top 10 cm of soil. Changes in the availability of the other plant

Table 2. Analyses of forest soils one year after application of alum sludge.

		Concentration in Soil Extract (mg/kg)							
Plot	**pH**	**NO_3^-**	**NH_4^+**	**P**	**K**	**Ca**	**Mg**	**Al**	**Mn**
Deciduous control									
litter[a]	5.3	6 M	12 L[b]	12 L	60 L	500L	75 H	10 L[b]	20 MH
0 - 5 cm	4.5	6 M	12 L	6 VL	30 VL	250 VL	15 L	125 H	5 L
5 - 10 cm	4.7	6 M	12 L	6 VL	30 VL	250 VL	12 L	125 H	5 L
Deciduous alum									
litter	6.2	5 M	12 L	12 L	60 L	500 L	50 MH	25 M	9 M
0 - 5 cm	4.8	6 M	12 L	6 VL	30 VL	250 VL	12 L	125 H	5 L
5 - 10 cm	5.0	6 M	12 L	6 VL	30 VL	250 VL	12 L	125 H	5 L
Coniferous control									
litter	4.6	3 L	12 L	9 L	45 L	425 L	35 M	20 M	10 M
0 - 5 cm	4.7	3 L	12 L	6 VL	30 VL	250 VL	20 ML	125 H	10 M
5 - 10 cm	4.8	3 L	12 L	6 VL	30 VL	250 VL	12 L	125 H	15 M
Coniferous alum									
litter	5.3	3 L	12 L	6 VL	30 VL	500 L	50 MH	50 MH	5 L
0 - 5 cm	5.1	3 L	12 L	9 L	45 L	425 L	25 M	125 H	5 L
5 - 10 cm	5.0	3 L	12 L	9 L	60 L	250 VL	20 M	125 H	5 L

VL = very low; L = low; ML = medium low; M = medium; MH = medium high; H = high; VH = very high

a - sample depth

b - low levels of these elements are optimum

nutrients shown in Table 2 appeared minimal.

Another way to determine effects of alum is to analyze growing plant tissue. Analyses of the deciduous leaves and coniferous needles are shown in Table 3. Since hemlocks retain their needles for several years, growth from the previous two years was analyzed.

There were only small differences in nutrients amongst treatments. Uptake of phosphorus appeared to be slightly less in the coniferous plots where alum was applied. Uptake of manganese was marginally less in the alum-treated plots of both forest types. Although no effects on growth were noted, the manganese relationship resembles the effects found in the greenhouse study of potting media amended with alum. Measurements on these plots will probably have to continue for many years before any long-term effects caused by the alum can be determined.

We draw these conclusions from the experiments on forested land:

- Alum can be applied to the forest floor from a tank truck.
- Little effect on the forest understory or the appearance of the forest floor was noticeable after 1170 m³/ha (124,800 gal/acre) of alum, containing 1.5% solids, were applied in split applications.
- The most notable change in soil after application is an increase in pH of approximately 0.5 to 1.0 unit.
- Plant nutrient uptake, as measured by tissue analysis, appears to be largely unaffected by alum.

Table 3. Analyses of plant tissue one year after alum application.

	Nutrient				
	%		mg/gm		
Treatment	**P**	**K**	**Ca**	**Mg**	**Mn**
Deciduous Control	0.19	0.5	14.7	1.7	1.8
Deciduous Alum	0.19	0.5	15.1	1.7	1.5
Hemlock Control 1 yr.	0.20	0.4	5.7	1.7	1.5
Hemlock Alum 1 yr.	0.19	0.4	5.6	1.7	1.2
Hemlock Control 2 yr.	0.17	0.2	6.5	1.6	1.8
Hemlock Alum 2 yr.	0.15	0.3	6.4	1.7	1.5

Conclusions

We found that dried alum improved the physical properties of potting media and acted as a liming material. The growth of plants, however, was restricted by phosphorus deficiencies induced by the ability of the alum to adsorb phosphorus in fertilizer and convert it into forms unavailable for plant growth. No toxic effects of the sludge or any of its constituents were observed.

Liquid alum sludge containing 1.5% solids sprayed on deciduous and coniferous forest plots at the rate of approximately 1170 m³/ha (124,800 gal/acre) increased soil pH by about 0.5 to 1.0, but had no effect on the nutrition or growth of the trees. As this rate was about one-tenth that used in experiments with potting media, it is unlikely that the availability of phosphorus will be reduced.

Because the most striking feature of alum sludge is its ability to adsorb phosphorous from soil and water, it might play a role in the removal of phosphorus in the effluent from sewage-treatment plants. Although the U.S. Environmental Protection Agency has banned the former practice of discharging alum sludge to surface waters, this decision might be reexamined in light of the increased attention given to alum treatment of eutrophic lakes.

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