PROPYLENE GLYCOL AND ETHANOL AS A REPLACEMENT ANTIFREEZE FOR PRECIPITATION GAUGES: DILUTION, DISPOSAL, AND SAFETY

by

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THE PROBLEM

Many agencies and organizations collect precipitation information by operating weighing bucket or storage precipitation gauges. At remote sites in cold climates, antifreeze is necessary to protect the gauge and to melt solid precipitation. Without melting, the snowfall from a single storm could overtop the gauge's bucket, or the snow from several storms could fill a storage gauge. The antifreeze must meet a range of criteria: it should be concentrated enough so that it can absorb several times its volume without freezing during the coldest temperatures, and it must be self-mixing so that ice does not form at the top of the fluid. Also, the admixed materials should be reasonably economical to obtain and dispose of, and the antifreeze should not pose a hazard to users or the environment when used in the designated manner.

The antifreeze that is currently used by many agencies is a mixture of ethylene glycol (used in automotive antifreeze) and methanol, also known as glycometh. The typical proportion is 40% ethylene glycol (EG) and 60% methanol. Between 250 and 1000 ml (8 to 32 oz) of mineral oil is typically added to the top of the antifreeze to suppress evaporation of the methanol and water during periods without precipitation. The specific gravity of the solution ranges from 0.94 to nearly 1.00 over the typical dilution range, so rain or melting snow sinks and keeps the solution mixed (Mayo, 1972).

Although EG has many positive attributes, it has one significant problem that is causing agencies to search for a substitute fluid: it is a toxic material even in its water-diluted form. In its pure form, the dose that is lethal to 50% of mammals (LD_{50}) is 1.4 ml/kg (Merck, 1976). Methanol is also toxic and flammable, but it evaporates quickly if exposed to air, so its toxicity is less of an issue than that of EG. It is no longer considered acceptable to dump glycometh on the ground near a gauge site, so it must be removed from the site by the gauge operator. Once removed to a central site, it must be picked up by a U.S. Environmental Protection Agency-approved waste hauler and recycled or disposed of in an acceptable manner. The mixture of water, EG, methanol, and oil is not simple to distill or refine, so disposal costs are high: between $1 and $4 per liter ($5 to $20 per gallon). The typical 300-mm (12-inch) dual traverse precipitation gauge yields 8-10 liters (~2 gallons) per charge change, and an annual storage gauge can produce up to 400 liters (~100 gallons), depending on the site's annual average precipitation amount. Some agencies, such as the National Weather Service and the U.S. Department of Agriculture's Soil Conservation Service and Forest Service, operate hundreds of gauges throughout the U.S., so the volume, cost and potential environmental risk of glycometh disposal are large enough that new procedures must be developed.

Recent research has identified propylene glycol (PG) and ethanol as reasonable substitutes to replace glycometh (U.S.D.A., 1989; Yeaman and Snyder, 1989). PG is used as an antifreeze in breweries and dairies, as a solvent in food colors and flavors, and as an airplane deicer. Ethanol (anhydrous alcohol) is the intoxicating agent in alcoholic beverages, and is widely used as a solvent in laboratories and industrial processing. Denaturing agents (methanol is commonly used at 5% by volume and other solvents may also be

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present to add odor) are added to industrial supplies to render the ethanol unpalatable without significantly affecting its properties or disposal characteristics. Ethanol evaporates rapidly when not contained and is flammable. A PG-ethanol (PGE) mixture has somewhat different density and freezing properties than the well-understood glycometh solution. The density of PG is 1.04 kg/l at 20°C, and the density of ethanol ranges from 0.84 to 0.79 kg/l at 20°C as the fraction of water decreases from 20% to near 0% (Weast, 1977). To mimic the self-mixing attributes of glycometh, the specific gravity of the PGE mixture should be between 0.93 and 0.99 from concentrate to full dilution at normal temperatures. The effect of low temperature on the specific gravity of the ratios of PGE over a range of dilutions with water is not known.

This paper reports on the freezing point and specific gravity of various ratios and dilutions of PG and ethanol. In addition, the transport and fate of the solution in the environment must be documented before it is used in precipitation gauges. Further, safety and health considerations will be addressed.

METHODS

PGE Ratios and Test Materials

Five ratios of PG-to-ethanol were evaluated, ranging from 20:80 to 60:40. Two 1-liter bottles were filled with 675 ml of solution for each of the PG:ethanol ratios (Table 1). The PG was obtained from JT Baker Company of Phillipsburg, NJ, catalog number 9402-03 (U.S.P grade, $137 per 4-liter bottle).* The PG formulation is 99.5% pure PG by weight, with the balance being water. Industrial grades of PG are 99% pure and cost $1.75 per liter when purchased in bulk. The ethanol was obtained from EM Science of Gibbstown, NJ, catalog number EX0285-3 ($58 per 4-liter bottle). The alcohol was 92% ethyl alcohol, 5% methanol, 1% ethyl acetate, 1% methyl-iso-butyl-ketone, and 1% hydrocarbon solvents.

Temperature Range and Dilutions

The winter temperatures that gauges are exposed to vary regionally, but precipitation rarely occurs at temperatures much below -35°C (Yeaman and Snyder 1989). This study evaluated five temperatures between 0 and -40°C in 10-degree increments. After a considerable search within the United States, cold rooms capable of -55°C were located at Environment Canada's National Hydrologic Research Institute (NHRI) in Saskatoon, Saskatchewan. The controls in the cold room were set to the desired temperature, and measurements were made between one-half and 1 hour after the room had reached the desired temperature. Solution temperatures were measured with mercury thermometers (ASTM #114C, gradated at 0.5°C, -80 to +20°C) in about 25% of the bottles.

Table 1

VOLUMES OF PROPYLENE GLYCOL (PG) AND ETHANOL FOR EACH SET OF DILUTIONS

Ratio (%) PG : Ethanol	Volume (ml) PG	Ethanol
20 : 80	135	540
30 : 70	203	472
40 : 60	270	405
50 : 50	338	338
60 : 40	405	270

* Mention of commercial products and organizations in this report does not constitute endorsement by the USDA over other products and organizations not mentioned.

Trays of 250-ml wide-mouth Nalgene bottles were arranged in a 5x10 matrix analogous to the five ratios and ten dilutions. Dilutions that were tested were: 1:0, 1:0.33, 1:0.66, 1:1, 1:1.5, 1:2, 1:3, 1:4, 1:5, and 1:7. The PGE solution was measured into the 250-ml Nalgene bottles using an appropriate size of volumetric cylinder and mixed with the required amount of water to achieve the desired dilution (Table 2). Each bottle was filled with about 150 ml of solution, and two 5x10 matrices were mixed separately to isolate mixing errors.

Table 2

VOLUMES OF EACH RATIO OF PROPYLENE GLYCOL AND ETHANOL SOLUTION (PGE) MIXED WITH WATER TO ACHIEVE TEN DILUTIONS SIMULATING MIXING WITH PRECIPITATION

Dilution PGE:Water	Volume (ml) PGE	Volume (ml) Water
1 : 0	150	0
1 : 0.33	113	37
1 : 0.66	90	60
1 : 1	75	75
1 : 1.5	60	90
1 : 2	50	100
1 : 3	38	112
1 : 4	30	125
1 : 5	25	125
1 : 7	19	131

Cold Room Procedures

Three states were identified in the bottles: liquid, slush, and solid. Liquid was distinguished from crystalline slush by the presence of crystals visible to the naked eye. The distinction between slush and solid was made on the basis of whether a thermometer could be inserted easily into the solution. The distinction between liquid and slush was quite clear because the crystals in the slush made the solution cloudy and then opaque as the freezing progressed. No temperatures were taken of the solid state because thermometers could not be inserted.

RESULTS AND DISCUSSION

At 0°C, no freezing was observed except for one bottle (60:40 PG-to-ethanol, one part PGE diluted with seven parts water, or 1:7) that became slushy. The slush formation resulted in an elevated temperature, a pattern that was repeated at most of the other temperatures. As a fluid freezes, the phase change liberates heat that is released to the surrounding container and air. Over an extended period or with the use of a cooling fluid other than air, the elevated slush temperatures might have decreased.

At -10°C, slush formed in all the bottles that had solutions that were more dilute than 1:3 (Figure 1). Temperatures of the liquids ranged from -11 to -9°C, and temperatures of the slush ranged from -10 to -5°C. At -20°C, slush formed in all the bottles that had solutions more dilute than 1:1.5. Temperatures of the liquids ranged from -18 to -21.5 °C, and temperatures of the slush ranged from -11 to -21.5°C. The widening range of slush temperatures may have been related to limitations of the cold room. The cold room in which the 0, -10, and -20°C experiments were performed was rated to -25°C and did not have as high a velocity of internal air circulation as the cold room that was used for the -30 and -40°C tests.

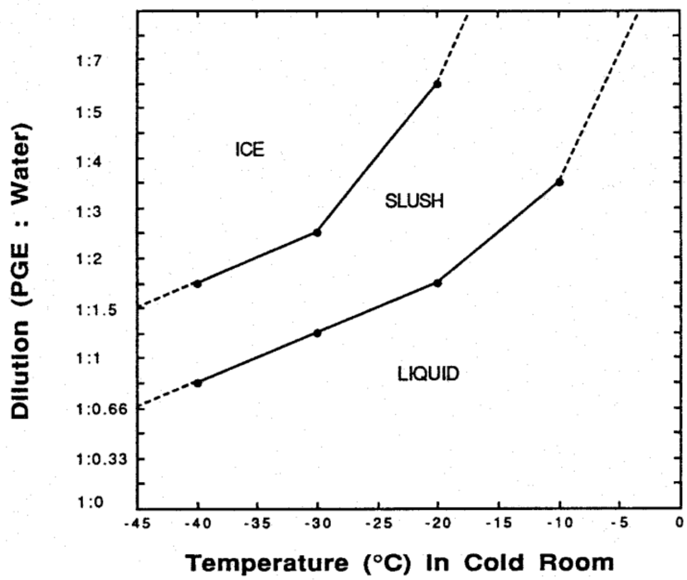

Figure 1. As temperature decreased, the more dilute solutions of propylene glycol and ethanol (PGE) progressed from liquid to slush or ice. Dashed lines are extrapolations of the cold-room data.

At -30°C, slush formed in the 1:1.5 and 1:2 dilutions, and ice was usually present at the 1:3 and higher dilutions (Figure 1). Temperatures of the slush ranged from -21 to -29.5°C. At -40°C, slush formed in the 1:1 and 1:1.5 dilutions, and ice formed in the 1:2 and higher dilutions. Temperatures of the slush ranged from -38 to -39.5°C.

On the basis of past work, there was an indication that the different PGE ratios might have different freezing characteristics (Mayo, 1972). The results from this experiment do not indicate any difference in freezing point that is not associated with dilution and temperature. Except for three cells in one of the two batches at -30°C, the other phase transitions are strongly associated with the steps associated with various dilutions. More intermediate dilutions might have exposed a different pattern, but the results from this experiment fit the needs of this study quite adequately. The selection of a PGE ratio to be used by agencies need not be influenced by concerns over different freezing characteristics of the various ratios.

Specific Gravity

Specific gravity (SG) is a measure of a material's density with respect to water, and the mixture of PG and ethanol has a specific gravity proportional to the relative proportion of the two materials in the mixture. When water is added to simulate the addition of precipitation to a gauge, the specific gravities of all three materials can be calculated as they change over the expected temperature range (Figures 2-3). A simple weighted average was used to estimate the specific gravity of the dilutions:

$$SG = [SG_{PGE} \times Ratio_{PGE}] + [SG_{water} \times Ratio_{water}] \qquad (1)$$

where the SG values are for each of the five temperatures and the ratios sum to one (Weast, 1977). Specific gravity values for -10, -20, and -30°C are intermediate between the SGs for 0 and -40°C that are shown in Figures 2-3. Past research used hydrometers to measure density directly, but hydrometers are difficult to use at low temperatures and the data are not reliable (U.S.D.A., 1989). The mixture must be less dense than water so that new precipitation sinks and promotes mixing in the gauge, but the solution must be more dense than the mineral oil (SG = 0.88 to 0.90) floating on the top that retards evaporation.

The mixtures having 20:80 and 30:70 ratios can be discounted for use as a precipitation gauge fluid because of their specific gravities at high temperatures (Figure 2). The 40:60 ratio is somewhat questionable as well because the SG of the solution at summertime temperatures might exceed the SG of the mineral oil and allow evaporation of the ethanol. The mixture having the 60:40 ratio has two drawbacks: it has such a high SG that self-mixing would be minimal, and the larger the amount of PG, the higher the cost. On the basis on these factors, the 50:50 solution appears to be the best choice when considering freezing and self-mixing concerns alone.

MIXING EQUATION FOR RECHARGING THE GAUGE

The U.S.D.A. Soil Conservation Service maintains a network of more than 400 precipitation gauging stations in the western United States. When each gauge is serviced during the summer, the appropriate amount of antifreeze must be added to melt the coming winter's snow and protect the gauge from freezing damage. The amount of antifreeze depends on a number of factors; chief among them are the average annual precipitation, the expected minimum air temperature, and the relative monthly percentage of the annual precipitation that can be expected. Other factors include the amount of fluid that is likely to be left in the gauge during servicing and expected deviation from the annual average precipitation. These factors are incorporated in the following equation (U.S.D.A., 1982):

$$\text{Recharge Quantity} = \frac{\left(\frac{(A*\text{Precip.}*B)+C}{\text{Max. \% Water in Solution}} - (A*\text{Precip.}*B)+C\right) - D}{E} \quad (2)$$

where

A = 1.2, typical deviation of 120% from normal precipitation,
B = 0.4, estimate that 40% of annual precipitation will have fallen by 1 January,
C = 11.7 cm (4.6 in.), estimated water content of fluid left in the gauge after draining, based on the assumption that the fluid is 80% water,
D = 3.1 cm (1.2 in.), estimated antifreeze content of fluid left in the gauge after draining, based on the assumption that the fluid is 20% antifreeze,
E = 1.3957 cm (2.0868 in. for gallons), conversion constant to transform volume to areal depth, based on assumption of a storage gauge with a 30.5 cm (12 in.) outer diameter, charged with 1 liter (or 1 U.S. gallon) of antifreeze solution,
Recharge Quantity = volume of concentrated PGE that is added at the time of service, in liters (divide by 3.785 for gallons),
Precip. = average annual precipitation for the site, in centimeters (inches), and
Max. % of Water in Solution = decimal percent of the maximum amount of water permitted in solution for the specified temperature so that freezing does not occur at that temperature.

In the 1982 version of the Soil Conservation Service's National Engineering Handbook, Section 22, the maximum percentages of water were specified at five temperatures, based on hydrometer and cold-room results using glycometh (Mayo, 1972). Based on the cold-room work reported above for propylene glycol and ethanol, new maximum water percentages have been estimated (Table 3). Users who have different conditions than those assumed by the selection of values for the variables A through E can calculate recharge quantities specific to their needs. In Figure 1, slush formed at different dilutions because of the varying temperature. The percent water associated with the slush/liquid line in Figure 1 is the maximum January 1 percent water in Table 3. These percentages reflect the midpoint between the dilution which remained liquid and the dilution that turned to slush. The midpoint was used to estimate a maximum water percentage, rather than the last liquid dilution percentage, so that less PGE is required to charge a gauge. Using less PGE saves money and leaves more room for precipitation in the gauge. Also, adding less PGE produces a less concentrated solution to be disposed of during summer. Occasional slush formation during times of extreme cold are unlikely to damage the gauge or cause capping over of the gauge, which would result in undermeasurement.

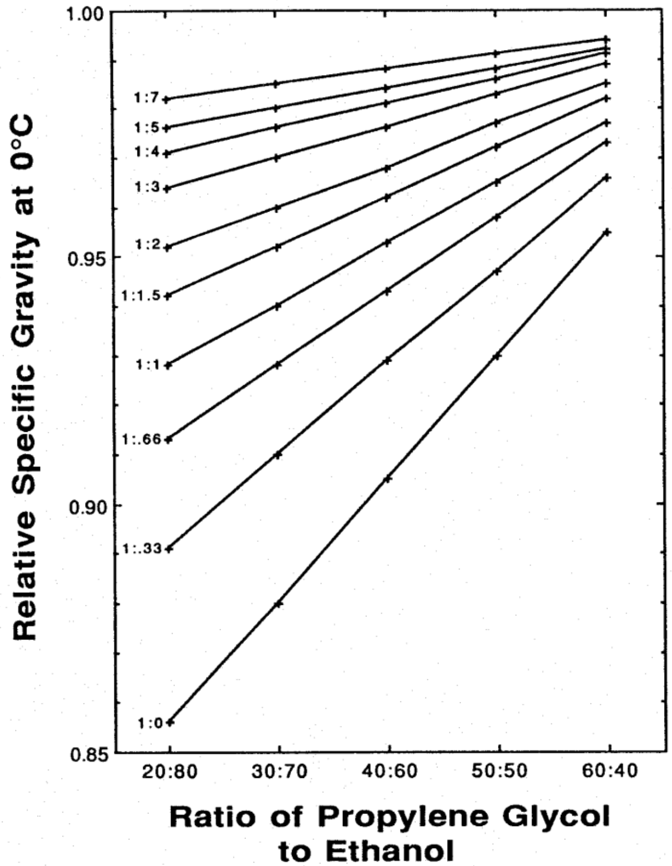

Figure 2. As solutions at 0°C increased from pure propylene glycol and ethanol to a dilution of 1:7, specific gravity increased from about 0.85 to 0.99.

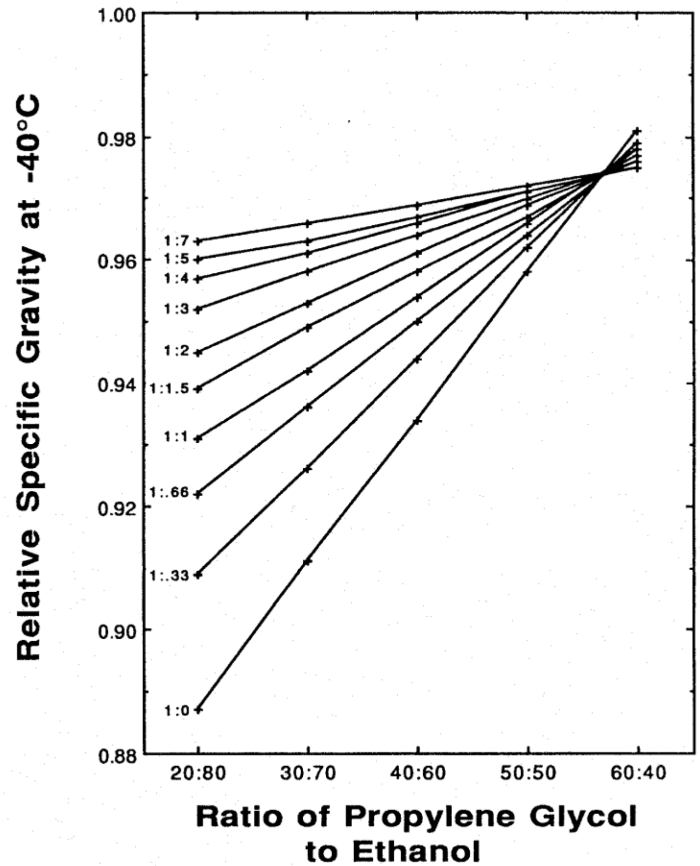

Figure 3. At -40°C solutions ranging from dilutions of 1:0 and 1:7 had specific gravities between 0.89 and 0.98; the lines cross because of the low specific gravity of water (0.974) at very cold temperatures.

Table 3

MINIMUM OCTOBER 1 RECHARGE VOLUMES OF A 50:50 MIXTURE OF PROPYLENE GLYCOL AND DENATURED ETHANOL FOR PRECIPITATION GAUGES, IN LITERS (U.S. GALLONS)

Minimum January 1 Temp. °C	°F	Maximum Jan. 1 Water %	Average Annual Precipitation: cm (inches) 51 (20)	76 (30)	102 (40)	127 (50)	152 (60)	178 (70)	203 (80)	229 (90)	254 (100)	305 (120)	530 (140)
-10	14	78	5.1 (1.3)	7.1 (2.0)	10@ (2.6)	12 (3.3)	15 (3.9)	17 (4.6)	20 (5.2)	22 (5.9)	25 (6.5)	30 (7.8)	35 (9.2)
-20	-4	64	12 (3.3)	17 (4.5)	22 (5.8)	27 (7.1)	32 (8.4)	37 (9.7)	42 (11.0)	47 (12.3)	52 (13.6)	62 (16.2)	72 (19.0)
-30	-22	56	18 (4.8)	25 (6.6)	32 (8.4)	39 (10.2)	46 (12.0)	52 (13.8)	59 (15.6)	66 (17.4)	73 (19.2)	87 (22.8)	101 (27)
-40	-40	45	29 (7.7)	40 (10.6)	51 (13.4)	61 (16.2)	72 (19.0)	83 (21.8)	94 (24.7)	104 (27.5)	115 (30.3)	136 (36.0)	157 (41)
-50	-58	35*	46 (12.1)	62 (16.3)	78 (20.6)	94 (25.0)	111 (29.2)	127 (33.5)	143 (37.8)	159 (42)	176 (46)	208 (55)	242 (64)

* : Estimated
@ : Volumes greater than 10 liters (40 gallons) rounded to nearest unit

ENVIRONMENTAL CONSIDERATIONS FOR DISPOSAL

The disposal problem is the primary motivation for incurring the increased costs of switching from glycometh to PGE. In that the desired disposal technique is discharge on the ground at the disposal site, three factors must be considered. The material must not be highly attractive and toxic to humans or animals if it pools at the site, and it must evaporate or be biodegradable by soil organisms. Finally, if transport across or within the soil allows PGE to reach a stream, it must not be particularly toxic to the aquatic ecosystem or be persistent. Each of these issues will be explored below.

Typical Disposal Conditions

Because of the guidelines for the siting of precipitation gauges, most gauges are at least midway up a basin and in a reasonably flat or gently sloping area. A few gauge sites may have minimal soil depth due to rocky outcrops, and some may be within a short distance of a stream that may or may not support fish. The storage-type gauges are typically visited during the summer and the prior winter's accumulated precipitation is drained out and the gauge is recharged with new antifreeze. Most gauges have a drain plug assembly, and the water/antifreeze fluid is drained into buckets or directly onto the ground. A storage gauge may contain 400 liters (~100 gallons) of fluid, of which between 15 and 50% could be the antifreeze solution. Assuming the antifreeze is equal parts PG and ethanol and that the gauge has the concentration of antifreeze that protects it to the minimum temperature of -40°C, 100 liters each of PG and ethanol would be discharged. In areas where moderate soil depths and cover conditions occur, the material would typically infiltrate into the soil within 30 minutes.

Of the three fluids, ethanol is the most volatile (vapor pressure temperature of 78.4°C at 760 mm Hg) and will evaporate rapidly under typical summer conditions (Weast, 1977). Water is the second most volatile (vapor pressure temperature of 100°C at 760 mm Hg), and some of the water would certainly evaporate, but the majority would infiltrate into the soil. Propylene glycol is the least volatile (vapor pressure temperature of 214.2°C at 760 mm Hg), and little is expected to evaporate.

Mammalian Toxicity

Propylene glycol has an LD_{50} of 25 g/kg in rats, and pure ethanol has an LD_{50} of 13.8 g/kg in rats, so the toxicity of both chemicals is relatively low (Merck, 1976). Because PG is not considered to pose a health hazard for humans, it is used in foods, dyes, and water systems. In laboratory tests, a variety of oral doses for humans and animals produced various mild and temporary reactions. Skin exposure produced no irritation under open or occluded conditions, but reddening and swelling were observed in one case in which applications were covered (occluded) to maximize effects. PG in its concentrated form may irritate the eyes but is unlikely to cause permanent damage (Hazardous Substances Database, 1991). Because of the potential irritation to the eyes from both PG and ethanol, safety glasses should be worn by anyone who is pouring these materials.

Ethanol is widely used in the medical and industrial fields as a sterilant and solvent. The denatured form that would be used in an antifreeze fluid will have methanol, methyl-iso-butyl ketone, or ethyl acetate added to discourage ingestion. Each of the additives causes different adverse effects to humans, and ingestion of significant quantities of denatured ethanol can cause blindness, severe stomach pain, and even death, although such an event is unlikely due to the noxious taste and odor caused by the denaturants.

Soil Degradation

PG is released into the environment during its commercial production and after its various uses such as a deicing fluid, as a solvent in food colors and flavors, as a lubricant or heat-transfer fluid during food processing, and as an antifreeze in the plumbing of recreational vehicles and boats. PG has been shown to biodegrade readily in both aerobic and anaerobic cultures typically used in water treatment systems (Kaplan, et al, 1982). In experiments with PG in active cultures in aerobic nutrient broths, PG was not detectable after 2 days. In cultures in which PG was the sole carbon source, it disappeared after 4 days under aerobic and 9 days under anaerobic conditions (Kaplan, et al, 1982). Because of the rapid microbial degradation, PG is expected to degrade rapidly under typical summer temperatures in soil that has a normal complement of microbial life. However, PG is completely soluble in water and has a low log Kow coefficient (-0.92), so it can be expected to be susceptible to significant leaching (Hazardous Substances Database, 1991). For leaching to occur, however, an ample supply of water must be applied in the week or two before biodegradation occurs. In much of the western United States, copious summertime precipitation is rare.

The ethanol in the dilute antifreeze solution may kill most of the soil microbes and some of the soil insects in the top 2-10 cm of soil when the solution is poured on the ground. The loss will be short term because of rapid microbial recolonization of the area. Both the PG and the alcohol will become nutrients for recolonizing microbes and insect life. Within 2 weeks of the application, sampling of microbes in the soil would be unlikely to show any residual effect (Poth, 1992).

Aquatic Effects

Although PG has been detected in wastewater treatment effluent streams, little is known about its toxicity to aquatic species (Hazardous Substances Database, 1991). In one aquatic test of a marking fluid for roosting blackbirds, a solution was tested for acute toxicity with four common aquatic species (Bills and Knittle, 1986). The solution was 25% PG, 25% water, ~48% Carboset (a resin to cause hardening), and trace amounts of fluorescein, emulsifiers, and colloids. Fathead minnows, channel catfish, bluegills, and rainbow trout were exposed to the chemical at concentrations ranging from 25 to 6,000 parts per million (ppm) for up to 96 hours. The compound was found to be essentially non-toxic. None of the fathead minnows or channel catfish died, and only 10% of the rainbow trout and 30% of the bluegills died after 96 hours in water with the highest concentration.

In a hypothetical spill of 5% (5 liters) of the 1:1 mixture over 5 minutes into a stream carrying 28 liters per second (1 cubic foot per second), 16 ml of PG per second would enter the stream. Diluted by the flow volume, the concentration would not exceed 600 ppm, a level at which no effect was seen in the above study. While direct entry of the antifreeze solution into flowing water should certainly be avoided, the effect of both the ethanol (due to evaporation) and the PG should be minimal.

CONCLUSIONS

A propylene glycol-ethanol solution would make an excellent replacement antifreeze solution for the glycometh solution that is commonly used in precipitation gauges. The PG portion of the solution is nontoxic, biodegrades rapidly, and is minimally toxic to aquatic ecosystems. The ethanol portion of the solution is toxic to mammals because of the denaturants added to prevent ingestion. Ethanol evaporates and infiltrates rapidly, however, and should have no long-term effect when discharged to the environment near the gauge site. Discharge on areas with some soil depth is recommended, and direct discharge to flowing water should be avoided.

Cold-room results showed that five different ratios of PG to ethanol did not yield differences in freezing characteristics. Ten dilutions with water were evaluated, and as temperature dropped from 0 to -40°C, increasingly concentrated solutions turned to slush and then ice. At -10°C, the dilution that had one part PGE to three parts water remained liquid. At -20°C, a dilution of 1:1.5 was required to prevent freezing. At -30°C, equal parts of PGE and water prevented freezing. At -40°C, a dilution of 1:0.66 was required to prevent freezing. These results were incorporated into an equation and a table used by the Soil Conservation Service that specifies the amount of PGE that should be used to recharge gauges based on factors such as annual average precipitation, minimum January 1 temperature, and winter precipitation pattern.

Calculations of specific gravities of the five ratios of PG to ethanol and the ten dilutions with water provided the basis for excluding the 20:80, 30:70, and 40:60 ratios. At 0°C and warmer temperatures, the relatively low specific gravities of these ratios might permit the PGE material to rise above the mineral oil that is commonly used. This problem is especially acute in gauges serviced and recharged early in the summer in warm sites at low elevations. Without the mineral oil as a cap to the solution, the ethanol would evaporate at a much more rapid pace and produce gauge records with declining catch values during fair weather.

Because self-mixing is more rapid and propylene glycol is more expensive than ethanol, the 50:50 ratio is recommended over the 60:40 ratio of PG:ethanol for use by agencies who maintain precipitation gauges. The PGE solution is an excellent substitute for the highly toxic glycometh solution that has provided protection from freezing for gauges in the past.

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