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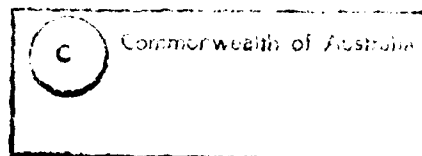
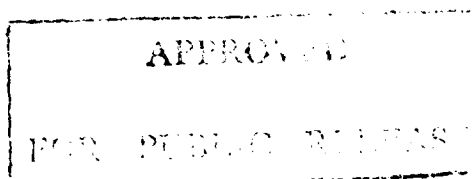
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Potential Replacements for Solvents
That Are Ozone Depleting
Substances

Lyn E. Fletcher

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Potential Replacements for Solvents That Are Ozone Depleting Substances

Lyn E. Fletcher

**Ship Structures and Materials Division
Aeronautical and Maritime Research Laboratory**

DSTO-TR-0046

ABSTRACT

The discovery of the hole in the ozone layer over Antarctica and the global drop in stratospheric ozone has prompted the banning of CFC-113 and 1,1,1-trichloroethane, which are common solvents used for degreasing and cleaning electronic components. The Royal Australian Navy uses these substances in a variety of applications and has initiated a replacement program. Two applications, cleaning baluns after lightning strikes and cleaning electrical motors have been selected as priorities under this program. This paper assesses the suitability of various solvents for these applications.

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Lyn Fletcher, BAppSc (Chem.) (RMIT) joined Materials Research Laboratory in 1985 and worked for three years on research into polymer and solvent interactions. She then joined a small multidisciplinary group investigating some environmental problems encountered by the Royal Australian Navy. In support of this work, Lyn is currently studying for a MEnvSc at Monash University.

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1. Introduction

The depletion of the ozone layer has resulted in international legislation banning various chlorohydrocarbons that remain in the atmosphere for a substantial time, thereby accumulating in the stratosphere and depleting the ozone layer. Two of these chemicals are the widely used cleaning solvents 1,1,1-trichloro-2,2,2-trifluoroethane (CFC-113) which has an atmospheric lifetime of 101 years and an ozone depleting potential of 0.86 (relative to CFC-11 = 1) and 1,1,1-trichloroethane, commonly called methylchloroform, which has an atmospheric lifetime of 6.3 years and an ozone depleting potential of 0.14, based on the University of Oslo's 2-D model calculations (Fisher *et al.* 1990). Since these substances are scheduled to be completely phased out of production by 1996 (Anon 1993), it is imperative that replacements be identified now.

The Royal Australian Navy (RAN) uses cleaning solvents in many dissimilar applications, all having different requirements for a cleaning solvent. To initiate the replacement program the RAN has identified two applications which are commonly carried out and have special requirements. The first is the cleaning of baluns which have been struck by lightning. A balun is a piece of electrical equipment that converts signal frequency. A lightning strike will cause carbonisation of the oil in the balun which then must be removed or shorting may occur. The second is the cleaning of electrical motors in both shore facilities and on board ship. Neither of these cleaning applications uses vapour degreasing equipment and the solvents are simply applied using a dip tank or by spraying.

Any replacement cleaning solvent must pose no health and safety risk to users, be non-flammable under the conditions in which it will be used and stored, be capable of removing the soil from the article to be cleaned while not affecting the underlying materials and must leave no residue to affect electrical contacts. In addition it is desirable that the cleaning solvent dry rapidly, be useable in existing cleaning equipment, have a long storage life and be inexpensive.

Possible replacement solvents include other chlorohydrocarbons and hydrochlorofluorocarbons with shorter atmospheric lifetimes that are not scheduled to be phased out of production, such as methylene chloride, trichloroethylene, perchloroethylene and 1,2-dichloro-1-fluoroethane; terpene based solvents which are derived from natural products (citrus or pine) and are generally blends; hydrocarbon blends which are formulations of higher molecular weight hydrocarbons that have a flash point above 50°C, and surfactant based formulations for use with water. In addition solvents have been formulated that combine the above types.

This paper will assess some of the non-aqueous solvents commercially available in Australia and will cover only selected examples of the many hydrocarbon and terpene blends available.

2. Description of Cleaning Applications

2.1 Baluns

Baluns are used extensively at Humpty Doo, NT by HMAS Coonawarra. In this climate lightning strikes of baluns are not uncommon. These strikes are unpredictable so the amount of affected equipment varies each year. The power rating of the Radio Frequency Systems T40K series baluns at Humpty Doo is 40 kW (80 kW Peak). The materials within the balun include polytetrafluoroethylene (PTFE), pressed fibreglass, solid non-jacketed copper wire with some Araldite present in earlier baluns. The Araldite was used in the manufacturing process to hold the coils together and is not considered critical to the performance of the equipment while the oil used in the baluns is a Shell diallyl oil (personal communication, David Bilton, RFS). Since any residue left within the balun has the potential to disrupt the electrical circuits and short the balun, any cleaning solvent used must leave no residue.

Previously affected baluns were disconnected and immersed in a 200 l drum of CFC-113 to remove the carbonised oil. When CFC-113 was first scheduled under the Montreal Protocol an alternative was sought. 1,1,1-Trichloroethane was considered to be the next best alternative. Unfortunately this has since been included on the list of substances to be phased-out of production. CFC-113 and 1,1,1-trichloroethane originally were selected as they leave no residue, have no effect on the materials in the baluns and pose minimal health and safety risk to users and can be stored for extended periods without the product deteriorating. Table 1 lists the properties of the solvents that are scheduled to be replaced.

Table 1: Physical and Chemical Properties of the Scheduled Solvents

	CFC-113 ¹	1,1,1-trichloroethane ^{1,2}
Appearance	Colourless liquid	Colourless liquid
Odour	Nil	Nil
Flashpoint, °C	Nil	Nil
Specific Gravity	1.579	1.338
Evaporation Residue, %		0.001
Acidity, %		0.001
Evaporation Rate ³ (perchloroethylene =1)	0.4	0.4
Water Content, mg/kg		0.01
Boiling Point, °C	46	72
Vapour Pressure, mmHg @ 20°C	300	100
Vapour Density, (Air=1)		4.6

1. Sigma-Aldrich (1993); 2. ICI (1992a); 3. Tested in this laboratory using method 121/86 in Def (Aust) 5631.

2.2 Electrical Motors

Cleaning electrical motors is a common use of 1,1,1-trichloroethane in the Navy and this is usually carried out at shore facilities particularly at HMAS Waterhen but in some instances electrical motors may need cleaning on board ship. The motors are sourced from a number of manufacturers and can include a wide variety of materials. The common materials encountered include ferrous metals and aluminium alloy for casings, copper wiring that can be jacketed with polyvinyl chloride (PVC), enamelled and varnished windings, silicone coated meter tails, adhesives, fibre insulation, cotton or fibreglass tape and lubricating oil and greases. These must be routinely cleaned to remove any degraded and carbonised grease, grit and dust that has entered through the motor casing. This is currently carried out using simple dip tanks, wiping with a solvent or spraying depending on the size and the position of the motor.

The Def (Aust) 5625a specification covered this application. It called for the use of narrow cut white spirit and 1,1,1-trichloroethane blends that had the chemical and physical characteristics listed in Table 2. Therefore, while the new replacements cannot meet the specification in regard to materials, any replacement should ideally meet the other physical and chemical requirements of the original specification.

Table 2: *Chemical and Physical Properties Required of a Cleaning Solvent used in Electrical Contact*

Physical Property	DEF(AUST) 5625A Specification
Flashpoint, °C	>50 (50% Residue)
Evaporation Residue, %	<0.01
Acidity, %	<0.001
Evaporation Rate (perchloroethylene = 1)	<5
Water Content, mg/kg	<100

3. Physical Properties of the Alternative Solvents

It is important that any new solvent be compatible with the conditions under which the cleaning is to be done. Tables 3, 4 and 5 list the chemical and physical properties of the potential replacements.

Table 3: Chemical and Physical Properties of Hydrocarbon based Cleaning Solvents

Physical Property	Evolve CH10 ¹	Evolve CH11 ¹	Evolve CH12 ¹	Axarel 6100 ²	Axarel 9100 ³
Manufacturer	ICI	ICI	ICI	Du Pont	Du Pont
Appearance	Clear, colourless liquid	Clear, colourless liquid	Clear, colourless liquid	Colourless liquid	Colourless liquid
Odour	None	Slight	Slight	Slight	Slight
Composition	Blend of Hydrocarbons	Hydrocarbons and Oxyhydrocarbons	Hydrocarbons and Oxyhydrocarbons	Aliphatic hydrocarbons and diisobutyl DBE ⁴	Aliphatic hydrocarbons and diisobutyl DBE ⁴
Flashpoint, °C	58	48	58	61	93.3
Specific Gravity				0.81	0.83
Evaporation Rate ⁵ (perchloroethylene = 1)	10		13	>25 ⁶	>25 ⁶
Surface Tension, mN/m	23	23	23		
Viscosity, mm ² /s	1.2	0.76	1.4		
Boiling Point, °C				182 - 295	221-295
Vapour Pressure, mmHg				<0.1	<0.1
Vapour Density, (Air=1)				6.7	6.7

1. ICI (date unknown); 2. Du Pont (1992a); 3. Du Pont (1992b); 4. Diisobutyl DBE is a mixture of Diisobutyl glutarate, diisobutyl adipate and diisobutyl succinate; 5. Tested in this laboratory using Method 121/86 in Def (Aust) 5631; 6. Difficult to dry completely since a virtually nonvolatile residual thin film remains over the test piece.

Table 4: Chemical and Physical Properties of Terpene based Cleaning Solvents

Physical Property	Citrex ¹	Citrasafe ²	Teksol EP ³	CN Solvent ⁴	Glidsafe UTS-48 ⁵	Glidsafe UTS-2 ⁶
Manufacturer	Callington Haven	Callington Haven	Callington Haven	Callington Haven	Glidco	Glidco
Appearance	Colourless liquid	Colourless liquid	Clear to Yellowish liquid	Colourless liquid	Colourless liquid	Colourless liquid
Odour	Slight amine	Citrus	Slight Citrus	Slight amine		
Composition	Terpenes, alkylpyrrolidones and glycol ether PNB	d-Limonene	C10-C11 hydrocarbons and d-limonene	d-Limonene and methylpyrrolidone	Dipropylene glycol monomethyl ether and terpenes	Terpenes and surfactants
Flashpoint, °C	55	54	44	53	50	48
Specific Gravity	0.86	1.84	0.77	1.11	0.86	0.882
Evaporation Rate ⁷ (perchloroethylene = 1)	>25	5			6	
Boiling Point, °C	171	171	155-171	171	151	105
Vapour Pressure, mmHg	<2	2	<10	<2		
Vapour Density, (Air=1)	4.5		>5.0	4.5	>1	>1

1. Callington (1993); 2. Callington (1992a); 3. Callington (1992b); 4. Callington (1992c); 5. Glidco (1991a); 6. Glidco (1991b); 7. Tested in this laboratory using Method 121/86 in Def (Aust) 5631.

Table 5: Chemical and Physical Properties of Chlorohydrocarbon Cleaning Solvents

Physical Property	Trichloroethylene ^{1,2}	Perchloroethylene ^{2,3}	Methylene chloride ^{2,4}	Genesolve 2000 ⁵
Manufacturer	Many	Many	Many	Allied Signals
Ozone Depletion Potential				0.11
Appearance	Colourless liquid	Colourless liquid	Colourless liquid	
Composition				1,2-dichloro-1-fluoroethane
Flashpoint, °C	Nil	Nil	Nil	Nil
Specific Gravity	1.460	1.623	1.330	1.24
Evaporation Rate ⁶ (perchloroethylene = 1)	0.6	1		0.1
Acidity, %	0.02	0.02	0.0005	
Water Content, mg/kg	0.01	0.01	0.01	
Surface Tension, mN/m				19.3
Boiling Point, °C	86-88	120-122	39-41	32.1
Vapour Pressure, mmHg @ 25°C	73 ⁷	19	435 ⁷	593
Vapour Density, (Air=1)		5.83		

1. ICI (1992b); 2. Sigma-Aldrich (1993); 3. ICI (1992c); 4. ICI (1992d); 5. Allied signals (date unknown); 6. Tested in this laboratory using Method 121/86 in Def (Aust) 5631; 7. Boublik et al (1984).

Pure methylene chloride and Genesolve 2000 (Table 5) can be disregarded as viable options for cold cleaning applications because of their low boiling points and high vapour pressures. Moreover, Genesolve 2000 has an ozone depleting potential (0.11) that is only slightly lower than 1,1,1-trichloroethane (0.14), so any benefit in converting to this alternative would be marginal, though at present it can still be manufactured until 2030 (Anon 1996). In addition, the lower boiling point and higher vapour pressure of Genesolve 2000 would result in greater losses to the atmosphere which would negate any benefit in reducing ozone depletion. Consequently this alternative will not be considered further. Trichloroethylene has a moderate vapour pressure and consequently significant concentrations could accumulate in the air over an open bath or tank.

4. Health and Safety Considerations

4.1 Toxicity

1,1,1-Trichloroethane has an occupational exposure limit - time weighted average in Australia (OEL TWA (Aust)) of 125 ppm, while CFC-113 has no OEL TWA (Aust) set and has had few toxicological studies performed (Sigma-Aldrich 1993). In comparison trichloroethylene has an OEL TWA (Aust) of 50 ppm and perchloroethylene has an OEL TWA (Aust) of 50 ppm (Sigma-Aldrich 1993). Consequently the potential replacements appear to be more toxic than those being replaced and will require stricter controls in their use to avoid breaching the OELs. The use of vapour control equipment is recommended if these cleaning solvents are adopted. The chlorohydrocarbon solvents therefore should not be used with spraying equipment

unless self-contained breathing apparatus is worn and all relevant safety procedures are strictly adhered to.

The hydrocarbon blends have no OELs set, however OSHA has set a personal exposure limit (PEL) of 400 ppm for mixed aliphatic hydrocarbons and Du Pont has set an acceptable exposure limit (AEL) of 1.5 ppm for diisobutyl DBE (Table 3). However, according to Du Pont no diisobutyl DBE has been detected over an open bath of either Axarel 6100 or Axarel 9100 (Du Pont 1992a & Du Pont 1992b). Therefore with the low vapour pressure of the hydrocarbon based solvents (Table 3) concentrations of these solvents in air should remain below their toxic level.

The terpene solvents are generally blends and since the composition of the terpene blends are not fully disclosed by the manufacturers it is not possible to accurately present toxicological data. However, no terpenes are listed as hazardous chemicals on OSHA's "Z" list, or listed by Section 303, of SARA title III or by section 302 of RCRA (Callington 1992d) and all have low vapour pressures (Table 4). Consequently these blends should pose no greater toxicological problems than other hydrocarbon solvents.

4.2 Flammability

CFC-113, 1,1,1-trichloroethane and the potential replacement hydrocarbons are non-flammable, whereas all of the hydrocarbon and terpene based solvents have flashpoints (closed cup) in the vicinity of 40-100°C. Def(Aust) 5625a mandates a flashpoint of greater than 50°C for the solvent remaining after 50% of it has been evaporated. Since this is for mixtures of 1,1,1-trichloroethane (which is non-flammable, has a low boiling point and a relatively high vapour pressure) and hydrocarbons, it is reasonable to expect 50% of the residue to include a large proportion of hydrocarbons. Consequently for the purposes of this report the figure of >50°C flashpoint will be considered for these cleaning solvents in their pure form. Using this value Evolve CH11, Teksol 44 and Glidsafe UTS-2 (Table 3 and 4) can be rejected as candidate replacements. It is also important for high pressure spray applications which can form aerosols thereby increasing the risk of fire, that the hydrocarbon and terpene blends should be used with caution. In such an application, using Axarel 9100 which has the highest flash point of 93.3°C (Table 3) would be preferable or the use of aqueous surfactant or terpene mixtures that are not covered by this report.

5. The Amount of Residue Left by the Alternatives

Since both applications involve electrical equipment and any residue left can disrupt electrical equipment and cause component failure, the amount of residue remaining after the cleaning solvent has dried is an important consideration. Def(Aust) 5625a calls for a residue of less than 0.01%. Most of the cleaning solvents do not have the residue levels mentioned in their technical specifications. This was tested at this laboratory using Def(Aust) 5631 method 120/86 and the results are listed in Table 6.

Table 6: Residue left after evaporation of the cleaning solvents

Cleaning Solvent	Residue, %
1,1,1-trichloroethane	0.001 ¹
Trichloroethylene	0.005 ¹
Perchloroethylene	0.005 ¹
Methylene chloride	0.001 ¹
Axarel 9100	0.07
Evolve CH10	0.005
Evolve CH12	0.01
Glidsafe UTS-4B	0.01
Citrasafe	2.3 ²
Citrex	0.1 ²

1. Results obtained from Manufacturers.
2. Oxidation and polymerisation observed.

Citrasafe, Citrex and Axarel 9100 do not meet the requirements for residue specified in Def (Aust) 5625A (Table 2). The chlorinated hydrocarbons and Evolve CH10 are similar to 1,1,1-trichloroethane and so should be acceptable for applications where low residue is very important. Glidsafe UTS-4B has antioxidant additives to stabilise the terpene blend which will account for the better performance of this product when compared to the other non-stabilised terpene blends, Citrasafe and Citrex.

It is important to note here that this work was restricted to using fresh cleaning solvents and cleaning solvents subject to oxidation and polymerisation such as hydrocarbons and terpenes could accumulate more residue with time. No attempt has been made to determine whether these levels could be significant.

6. Materials Compatibility of the Alternatives

Polymeric materials such as rubbers, plastics, resins and paints are generally the most affected by the presence of organic solvents. An incorrect choice of cleaning solvent can result in swelling, cracking, crazing, loss of mechanical strength and dissolution of the polymeric material, which could ultimately cause equipment failure. Therefore it is important that this is considered before any new solvent be used. There are two methods for assessing what effect a cleaning solvent will have on materials. The first is by direct testing and the second is a method based on comparing the physicochemical properties of the material and cleaning solvent using Hildebrand solubility parameters.

6.1 Direct Testing

Polymers can be affected by cleaning solvents in numerous ways. For a rubber or elastomer the cleaning solvents may cause solvation or swelling (depending on the amount of crosslinking in the polymer), while plastics can undergo cracking and crazing following contact with certain cleaning solvents.

However, of the solvents listed in Tables 3-5, very few have been subjected to this type of examination, or if tested, reported in the literature. The manufacturer Glidco (1993) has performed tests on Glidsafe UTS-4B and these are presented in Table 7. From this data it is observed that Glidsafe UTS-4B is not acceptable for use with all materials. Rubbers such as latex (natural rubber) and neoprene are incompatible with it, as are polystyrene and polyurethane which are often present as foams. Glidsafe UTS-4B should therefore be used with care when foams or rubber material are present.

Table 7: Results of Material Compatibility Testing for Glidsafe UTS-4B¹

Polymer	Glidsafe UTS-4B ²
Buna N (Nitrile)	F
CPVC	G
Delrin	E
Latex	N
Neoprene	N
Nylon	E
PETE	G
Polycarbonate	F
Polyethylene (HDPE)	G
Polyethylene (LDPE)	F
Polypropylene	F
Polystyrene	N
Polyurethane	N
PTFE	E
PVC	G
Silicone	F
Teflon	E
Viton	G

1. Glidco (1993):
2. E No damage after 7 days exposure at 25°C;
- G Little or no damage after 7 days exposure at 25°C;
- F Some effect after 7 days of exposure at 25°C (cracking, crazing, loss of strength, discoloration, softening or swelling);
- N Not recommended for continuous use. Immediate damage may occur

6.2 Hildebrand Solubility Parameters

The experimentation required to assess the direct effect of a cleaning solvent on a polymer is time consuming. Methods based on determining solubility parameters have been suggested and used to predict the behaviour of polymers in contact with different chemicals and these methods are widely used in the paint industry. In its

simplest form the Hildebrand solubility parameter (δ) is a function of the cohesive energy of the chemical which is related to the molar vaporisation energy ($\Delta_e U$) and the molar volume of the chemical (V) by equation 1 (Barton, 1983).

$$\delta = (\Delta_e U / V)^{1/2} \quad (1)$$

Table 8 lists the Hildebrand solubility parameters for the cleaning solvents and some closely related chemicals. This approach predicts that a chemical will be a solvent for a material if the Hildebrand solubility parameters for the two substances are the same. Therefore if the aim is to find a solvent to remove a specific material, the preferred solvent would have a Hildebrand solubility parameter that is closest to that of the dirt or soil to be removed.

Alternatively if the aim is to choose a cleaning solvent which is going to have the least effect on the underlying material, a cleaning solvent should be chosen that has a Hildebrand solubility parameter that is far removed from that of the material in question. It is not important whether the cleaning solvent has a Hildebrand solubility parameter that is smaller or larger than the polymer, as long as it is dissimilar.

Table 8: Hildebrand Solubility Parameters for Solvents at 25°C (based on values obtained by Hansen where available, and Gallagher)^{1,2}

Chemical Name	δ (MPa ^{1/2})
1,1,1-trichloro-2,2,2-trifluoroethane (CFC-113)	14.7
1,1,1-trichloroethane	17.7
Trichloroethylene	19.0
Perchloroethylene	20.3
Methylene chloride	17.0
Hexane	14.9
Octane	15.5
Decane	15.8
1-Octene	15.5
iso-Octane	14.3
cyclohexane	16.8
Benzene	18.4
d-Limonene	17.8
Glidsafe-UTS4B	18.2
Turpentine	16.5
isobutyl acetate	17.2
Diisobutyl phthalate	18.3
Dipropylene glycol monomethyl ether	19.0
Diethylene glycol monomethyl ether	22.3
N-Methyl pyrrolidone	23.0
Water	47.8

1. Barton (1983); 2. Gallagher (date unknown).

For blends, the Hildebrand solubility parameter can be determined by multiplying the volume fraction (ϕ) of the individual solvents with the individual solubility parameters and summing this for all the components. Consequently for a two component blend the solubility parameter is found by equation 2.

$$\delta = \phi_i \delta_i + \phi_j \delta_j \quad (2)$$

Therefore for hydrocarbon based solvents such as Axarel and Evolve and the terpene based solvents the Hildebrand solubility parameter can be estimated if enough data is available for each of the components. For example the Axarels contain aliphatic hydrocarbons and diisobutyl DBE (which is a mixture of diisobutyl glutarate, diisobutyl adipate and diisobutyl succinate) and from a consideration of the range of values of these type of compounds an estimate can be made. Table 8 shows that aliphatic hydrocarbons have solubility parameters that range from 14.9-16.8 MPa^{1/2} and iso-butyl acetate has a solubility parameter of 17.2 MPa^{1/2}. Isobutyl acetate was chosen to approximate diisobutyl DBE, since it is the most closely related chemical that has been quoted in the literature (Barton, 1983). Therefore it is expected that the Axarel blends should have Hildebrand solubility parameters that are somewhere within the range 14.9-17.2 MPa^{1/2}. It is impossible to be more accurate since the relative concentrations and identity of each component are unknown. This range should be relevant for all of the hydrocarbon blends listed in Table 3.

For the terpene blends not as many Hildebrand solubility parameters have been determined. However, terpenes are generally unsaturated hydrocarbons with limonene having a Hildebrand solubility parameter of 17.8 MPa^{1/2} and turpentine, the unpurified extract from pine trees having a Hildebrand solubility parameter of 16.5 MPa^{1/2}. Therefore the Hildebrand solubility parameters of the terpene blends are expected to be in the vicinity of 16.5-17.8 MPa^{1/2}. It must be recognised that some may contain other oxygenated compounds and chemicals with different functional groups (Table 4) which could raise the expected Hildebrand solubility parameters. For example, Glidsafe UTS-4B which is a mixture of terpenes and dipropylene glycol monomethyl ether has a Hildebrand solubility of 18.2 MPa^{1/2} (Gallagher date unknown). Citrasafe is solely limonene (Table 4) and therefore its Hildebrand solubility is 17.8 MPa^{1/2}.

Unfortunately the Hildebrand solubility parameters only work well for non-polar solvents. Polarity and other intermolecular interactions such as hydrogen bonding can alter the solubility so that a simple comparison of Hildebrand solubility parameters can lead to false results. Therefore, the Hildebrand solubility parameter has to be extended to take these interactions into account. One convenient and widely used method is the one proposed by Hansen (1967) which determines the proportion of the interactions that can be attributable to dispersion (δ_d), polarity (δ_p) and hydrogen bonding (δ_h) and are related to the Hildebrand solubility parameter by equation 3.

$$\delta^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \quad (3)$$

Using these values it is possible to do a three dimensional plot to determine the region where solvency occurs for a particular polymer and compare solvents to this region. Table 9 lists the Hansen parameters for the solvents of interest.

Table 9: Hansen Values for Selected Solvents at 25°C (based on values obtained by Hansen where available, and Gallagher)^{1,2}

Chemical Name	δ_d (MPa ^{1/2})	δ_p (MPa ^{1/2})	δ_h (MPa ^{1/2})
1,1,1-trichloro-2,2,2-trifluoroethane (CFC-113)	14.7	1.6	0
1,1,1-trichloroethane	17.0	4.3	2.1
Trichloroethylene	18.0	3.1	5.3
Perchloroethylene	19.0	6.5	2.9
Methylene chloride	15.3	6.1	3.9
Hexane	14.9	0	0
Octane	15.5	0	0
Decane	15.8	0	0
1-Octene	15.0	3.5	2.3
iso-Octane	14.3	0	0
cyclohexane	16.8	0	0.2
Benzene	18.4	0	2.0
Limonene	17.6	2.0	2.0
Glidsafe UTS-4B	17.4	3.1	4.5
Turpentine	16.4	1.4	0.4
isobutyl acetate	15.1	3.7	6.3
Diethylene glycol monoethyl ether	16.2	9.2	12.3
Water	15.6	16.0	42.3

1. Barton (1963); 2. Gallagher (date unknown).

Another variation has been proposed by Teas (1968) who devised a triangular plot based on fractions of the different Hansen parameters as defined by equations 4, 5 and 6. Where equation 4 is the fraction attributable to dispersion, equation 5 is the fraction attributable to polarity and equation 6 is the fraction attributable to hydrogen bonding.

$$f_d = \delta_d / (\delta_d + \delta_p + \delta_h) \quad (4)$$

$$f_p = \delta_p / (\delta_d + \delta_p + \delta_h) \quad (5)$$

$$f_h = \delta_h / (\delta_d + \delta_p + \delta_h) \quad (6)$$

Teas (1968) used this approach to determine the region for solvation for Vipla[®] KR polyvinyl chloride resin. When the solvents are included on the plot (Figure 1) it is observed that methylene chloride falls inside the region for solvents. Consequently this solvent should not be used when this resin is present. All the other chlorohydrocarbons occur at a similar distance from the solvent boundary and therefore trichloroethylene and perchloroethylene should have no greater effect on this polyvinyl chloride resin than 1,1,1-trichloroethane. The hydrocarbons and the terpenes are further removed again from the solvent boundary and should be acceptable for use with this material.

Figures 2 and 3 show the solvent boundaries for two other polymers (E-05 K epoxy resin and an alkyd resin which is commonly used in varnishes) that are likely to be present in electrical motors.

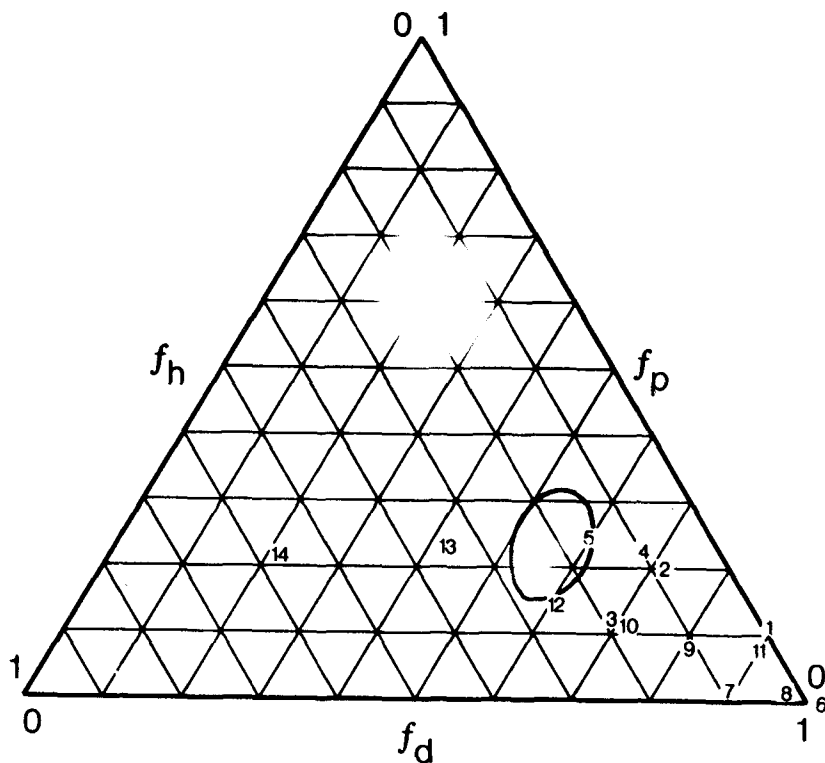


Figure 1: Triangular fractional solubility diagram for Vipla® KR poly(vinyl chloride) resin (Montecatini) (Teas 1968). 1. CFC-113, 2. 1,1,1-trichloroethane, 3. trichloroethylene, 4. perchloroethylene, 5. methylene chloride, 6. n-alkanes, 7. Benzene, 8. cyclohexane, 9. limonene, 10. Glidsafe UTS-4B, 11. turpentine, 12. iso-butyl acetate, 13. diethyleneglycol monoethyl ether, 14. water.

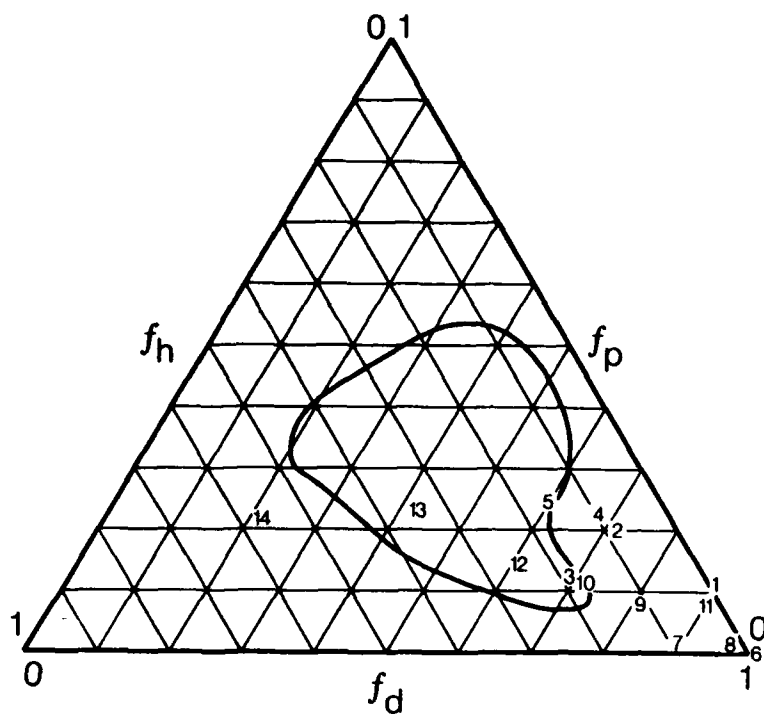


Figure 2: Triangular fractional cohesion parameter solubility diagram for E-05K epoxy resin (Loer & Lauruhcher in Barton 1983). 1. CFC-113, 2. 1,1,1-trichloroethane, 3. trichloroethylene, 4. perchloroethylene, 5. methylene chloride, 6. n-alkanes, 7. Benzene, 8. cyclohexane, 9. limonene, 10. Glidsafe UTS-4B, 11. turpentine, 12. iso-butyl acetate, 13. diethyleneglycol monoethyl ether, 14. water.

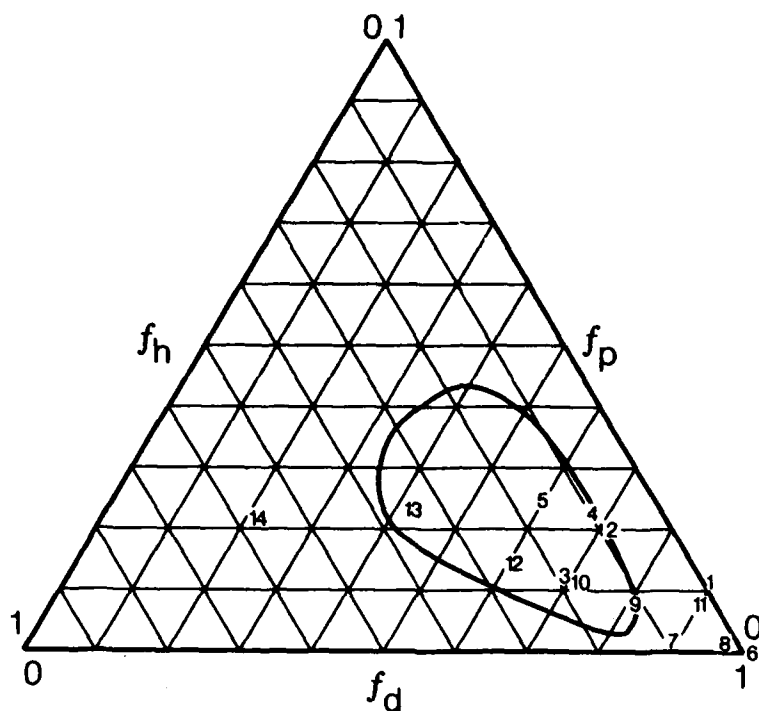


Figure 3: Triangular fractional cohesion solubility parameter diagram for an alkyd resin (Loer & Lauruhcher in Barton 1983). 1. CFC-113, 2. 1,1,1-trichloroethane, 3. trichloroethylene, 4. perchloroethylene, 5. methylene chloride, 6. n-alkanes, 7. Benzene, 8. cyclohexane, 9. limonene, 10. Glidsafe UTS-4B, 11. turpentine, 12. iso-butyl acetate, 13. diethyleneglycol monoethyl ether, 14. water.

Solvent boundaries however have not been determined using Hansen plots, triangulation or any other method for all polymers likely to be encountered. A method based on determining a radius of interaction (JR) has been suggested (Hansen 1967). This radius is determined by examining the behaviour of a polymer with various cleaning solvents and experimentally determining the best JR value that encloses the region where solvation or swelling occurs. This can then be used to predict the behaviour of other solvents by determining the radius between the specific cleaning solvent and polymer Hansen solubility parameters ($\bar{J}R$) and has been found to be 95% accurate at predicting the solubility of polymers in mixtures of chemicals (Hansen 1967). $\bar{J}R$ values are calculated using equation 7

$$\bar{J}R = (4(\delta_d - j\delta_d)^2 + (\delta_p - j\delta_p)^2 + (\delta_h - j\delta_h)^2)^{1/2} \quad (7)$$

where $(\delta_d, \delta_p, \delta_h)$ represent the Hansen solubility parameters for the cleaning solvent and $(j\delta_d, j\delta_p, j\delta_h)$ are the Hansen solubility parameters of the polymer. A substance will be predicted to be a solvent for the polymer when the calculated $\bar{J}R$ is less than the radius of interaction (JR). Subtracting the radius of interaction from the calculated radius for the polymer and cleaning solvent will consequently give an indication of polymer behaviour in the presence of a given cleaning solvent. A negative value is likely to indicate a solvent or swelling agent and should not be used for cleaning purposes. Cleaning solvents with slightly positive values should be used with caution but those with large positive numbers should have no effect or minimal effect on the polymeric material. However, it is important to note that this assumes a relatively evenly shaped solvent boundary and from Figures 1, 2 and 3 it is observed that solvent boundaries are not necessarily regularly shaped. Therefore this method can only be a useful guide to predicting polymer and cleaning solvent behaviour. Table 10 lists the resultant differences in radii ($\bar{J}R - JR$) for a number of specific commercial polymers and the solvents being considered as replacements for ozone depleting solvents.

Summing the differences in radii for a variety of polymers for a given cleaning solvent can act as a guide to determining which cleaning solvent will be less likely to affect any polymers adversely. The greater the number, the less likely is it that the cleaning solvent will create problems with polymeric materials in general. Using Table 10 it is observed that water has the greatest total, followed by the saturated hydrocarbons and CFC-113. The unsaturated hydrocarbons (terpenes, aromatics and unsaturated aliphatics) chlorohydrocarbons and oxygenated hydrocarbons have a greater probability of affecting an unknown polymeric material. Elastomeric materials such as natural and synthetic rubbers appear to be the most susceptible to the solvents considered and care should be taken whenever these materials are a component of the part being cleaned.

Unfortunately the above data applies to specific grades of polymers and variations will occur between manufacturers products and the grades of plastic, since all polymers are blends of materials with various molecular weights and incorporate additives such as plasticisers, antioxidants, dyes and fillers. Consequently one manufacturer's grade of polymer may have slightly different properties that could shift or enlarge the solvent boundary.

Table 11 shows the ranges of Hildebrand solubility parameters obtained for different manufacturer's grades of polymers. Therefore in situations where the exact solubility parameters of a particular grade of plastic are unknown the broader and less accurate comparisons of Hildebrand solubility parameters may be used in conjunction with the Hildebrand solubility parameters listed in Table 8. This will determine whether adverse interactions may occur and whether evaluation of the cleaning solvent with a test piece should be considered.

Table 11: Hildebrand Solubility Parameters for some Common Polymers in Contact with Solvents which have Poor Hydrogen Bonding Capability¹

Polymeric Material	δ (MPa ^{1/2})
Polytetrafluorocarbons (Teflon, PTFE)	12-13
Ester gum	14-22
Alkyd 45% Soy oil	14-22
Silicone DC-1107	14-19
Poly(butyl methacrylate)	15-23
Polyisobutylene	15-16
Polyethylene	16-17
Natural Rubber	17
Chlorinated Rubber	17-22
Polystyrene	17-22
Polyvinyl chloride	17-23
Phenolic Resins	17-24
Buna N (butadiene-acrylonitrile copolymer)	18-19
Polymethyl methacrylate	18-26
Polycarbonate	19-22
Polyurethane	20-21
Styrene-acrylonitrile copolymer	22-23
Vinsol (rosin derivative)	22-24
Epon [®] 1001 (epoxy)	22-24
Shellac	Not Soluble
Cellulose acetate	23-26
Cellulose nitrate	23-26
Polyvinyl alcohol	Not Soluble
Polyacrylonitrile	Not Soluble
Nylon 6, 6	Not Soluble
Cellulose	Not Soluble

1. Barton (1983)

Polytetrafluorocarbons such as PTFE have very low Hildebrand solubility parameters, therefore few chemicals are absorbed by this type of material. Perfluorocarbons are the most readily absorbed but these have Hildebrand solubility parameters that are extremely low (perfluoroheptane 11.9 MPa^{1/2}). Of the substances that are being considered for use as replacement solvents, the greatest absorption was found with perchloroethylene, which resulted in 1.9 % weight gain in a 2 mil thick PTFE film (Starkweather, 1977). Consequently the PTFE present in the baluns should not be significantly affected by any of the cleaning solvents being considered.

Many of the solvents have Hildebrand solubility parameters that overlap the ranges for the polymers mentioned in Table 11. The rubbers have generally lower and broader ranges (14-22) and are more likely to overlap the cleaning solvents. Therefore care should be taken when rubbers and elastomers are present in the components being cleaned. Highly polar and hydrogen bonding polymers such as cellulose are well above the range of the candidate solvents and should therefore not be affected.

Table 11 also serves to illustrate the great range of values that can occur between different grades of plastics. This is a major limitation to predicting the behaviour of polymers in the presence of solvents, whether the method of prediction is by direct examination as in the case of Glidsafe UTS-4B (Table 6) or by using solubility parameters.

6.3 Comparison of methods

Two methods have been proposed: direct testing and prediction using Hildebrand solubility parameters. The results obtained with Glidsafe UTS-4B can be compared with the results predicted in Table 10 and 11.

Table 12: Comparison of Results of Direct Testing and Hildebrand Solubility Parameters

Polymer	Glidsafe UTS-4	($\delta_H - \delta_R$)	Range δ
Latex (natural rubber)	N		17
Neoprene (Chloroprene)	N		
Polystyrene	N	-4.4	17-22
Polyurethane	N		20-21
Buna N (Nitrile)	F	-3.4	18-19
Polycarbonate	F		19-22
Silicone	F		17-19
Polyethylene (LDPE)	F		16-17
Polypropylene	F		
Polyethylene (HDPE)	G		16-17
PETE	G		19-22
CPVC	G		
PVC	G	2.5	17-23
Viton (Vinylidene fluoride - hexafluoropropylene copolymer)	G		
Nylon	E		not soluble
Delrin (Polyacetal)	E		
PTFE	E		12-13
Teflon	E		12-13

Only a few of the polymers tested for compatibility with the solvents have had their radius of interaction determined and these are listed in Table 10. However, of those that are listed there is reasonable correlation with the negative ($\delta_H - \delta_R$) values being not suitable or having some effect after 7 days, while the positive values were found to have little or no effect after 7 days. Unfortunately the small number of similar materials assessed using these methods limits the usefulness of this comparison.

Comparing the range of Hildebrand solubility parameters shows that the polymers with Hildebrand solubility parameters in the range 17-19 MPa^{1/2} are generally affected by Glidsafe UTS-4B. This contains dipropyleneglycol monomethyl ether and terpenes and has a Hildebrand solubility parameter of 18.2 MPa^{1/2}. This corresponds well with the above observation, except for polyurethane which has a Hildebrand solubility parameter of 20-21 MPa^{1/2}. In addition the triangular fractional solubility diagrams (Figures 1, 2 and 3) show that Glidsafe UTS-4B is a solvent for alkyd resin, border line with epoxy resin and is not expected to be a solvent for PVC. Only PVC has been directly tested and the results are in agreement. However, from Table 11 the epoxy resin (Epon 1001) is given a Hildebrand solubility parameter of 22-24 MPa^{1/2} which is well above the value of 18.2 MPa^{1/2} for Glidsafe UTS-4B. This illustrates that the solubility parameter approach is useful but when using a new solvent, caution should be exercised in case unexpected interactions occur.

7. Ability of the Solvents to Remove Grease and Oils

As mentioned in the previous section a solvent is one that has the chemical properties similar to the dirt or soil being removed. Both applications being considered are basically degreasing operations. Therefore solvents should ideally have Hildebrand solubility parameters that match the oil or grease being removed. The Hildebrand solubility parameters for some oils are listed in Table 13.

Table 13: Hildebrand Solubility Parameters of Oils¹

Substance	δ (MPa ^{1/2})
iso-Octane	14.3
ASTM fuel B	15.3
ASTM fuel C	16.3
ASTM oil #1	13.9
ASTM oil #2	15.6
ASTM oil #3	16.6
Brake fluid	19.8
Auto transmission fluid	14.3
Linseed oil	14.8
MIL-L-7808 (ester)	14.9
MIL-H-8446 (silicate)	17.5
MIL-H-5606 (Petroleum)	14.7
Motor oil SAE 20W	14.7
Phosphate hydraulic	18.4
Sperm oil	14.7
Castor oil	18.2
Linseed oil (white refined)	14.9
Mineral oil (white refined)	14.5
Pine oil	16.6
Cottonseed oil	14.9
Neats foot oil	15.1

1. Barton (1983).

Most of these oils and greases have Hildebrand solubility parameters in the range of 14-18.5 MPa^{1/2}, which is the general range for the cleaning solvents being assessed in this paper. For hydrocarbon based oils the Hildebrand solubility parameters are at the lower end of this range, while other synthetic and natural oils with different chemical structures and functional groups the Hildebrand solubility parameters are higher. This can be a useful guide to aid in choosing the appropriate solvent to remove different oils. Hydrocarbon solvents such as blends and terpenes have lower solubility parameters that should be better at removing hydrocarbon oils while the chlorohydrocarbons (such as perchloroethylene and trichloroethylene) have higher solubility parameters so they should efficiently remove other synthetic and more oxygenated and degraded oils. Therefore the more oxygenated and degraded the oil the higher the Hildebrand solubility parameter the solvent needs to be to effectively remove the oil or grease. Alternatively the solvent should be more polar or more capable of forming hydrogen bonds which is achieved by blending more oxygenated solvents with hydrocarbons or terpenes.

8. Discussion

8.1 Baluns

For cleaning Baluns after a lightning strike the amount of residue left is critical and therefore the solvent should be sufficiently volatile so that the solvent completely drains and dries from the balun after cleaning. The chlorohydrocarbons, such as perchloroethylene and trichloroethylene appear to be the best choice. However, the greater toxicity of these substances compared to 1,1,1-trichloroethane or CFC-113 will require stricter handling, storage and use procedures. Perchloroethylene has a lower vapour pressure than trichloroethylene and its vapour is therefore less likely to accumulate above a bath at concentrations that exceed the OEL TWA (Aust) of 50 ppm. However, vapour controlled baths are still considered the safest option to ensure that this limit is never exceeded when using trichloroethylene and perchloroethylene.

Neither trichloroethylene nor perchloroethylene should interact with the PTFE present in the baluns but the epoxy resin (Araldite) used in the manufacturing process of older baluns could be affected (Table 10, Figure 2). From Figure 2, perchloroethylene appears to have a lower likelihood of affecting epoxy resins (E-05K), though from Table 10 both solvents are predicted to be inside the region of interaction for Epikote 1001 epoxy resin, highlighting that variations in grades of epoxy resins will influence the outcome. Therefore, direct testing should be performed on a sample of the epoxy resin used, though some interaction could be tolerated because the epoxy resin is not critical to balun performance (personal communication, David Bilton, RFS). In addition the higher Hildebrand solubility parameter of perchloroethylene means that this cleaning solvent may be less effective at removing the diallyl oil present in the older style baluns but this will depend on the amount of degradation the oil has experienced. The Hildebrand solubility parameter of the diallyl oil is unknown but is probably going to be approximately 16 MPa^{1/2}, since its main components are

purified. However, both Citrasafe and Citrex resulted in large residues with the methods used for Def (Aust) 5625a and so care should be taken that during the drying process no oxidation of the solvent occurs. If parts can be wiped dry this will reduce drying times and reduce the likelihood of oxidation occurring but drying ovens should be avoided. Glidsafe UTS-4B did not leave large residues with the method used, so it could be used in conjunction with drying ovens. Glidsafe UTS-4B is composed of a blend of terpenes and dipropylene glycol monomethyl ether and has a Hildebrand solubility parameter of $18.2 \text{ MPa}^{1/2}$. This solvent should therefore be appropriate for removing many oils but is expected to have some effect on rubbers, polystyrene, polyurethane, polycarbonate and hydrocarbon and resins.

The solubility map for PVC is shown in Figure 2 and from this both the terpenes and hydrocarbons are outside the solubility range. The material included in the varnishes is unknown because of the large variety in use, however common varnishes are of the alkyd type. Figure 3 is a solubility map for an alkyd resin and it shows that the hydrocarbons are outside this region and the terpenes are either inside or close to the solubility range. Blending hydrocarbons with oxyhydrocarbons such as diisobutyl DBE or dipropylene glycol monomethyl ether could alter the characteristics of the blend and reduce the fraction attributable to dispersion, which could, depending on the proportion of the components, shift the solvent into the solubility range. Therefore Evolve CH10 which is composed solely of hydrocarbons is less likely to affect the alkyd resins commonly used in varnishes for winding than the other hydrocarbon or terpene blends.

The motors can contain rubbers (type unknown) that will likely swell in most of the cleaning solvents. It has been found that 1,1,1-trichloroethane will be absorbed into rubbers and can remain in the rubber for over 4 days (Purvis & Moran, date unknown). It has also been observed that rubber will return to normal once dry and that the evaporation rate of the cleaning solvent in air is not important because once the solvent is absorbed into the rubber, its desorption is dispersion limited (Purvis & Moran, date unknown). Therefore, the replacement cleaning solvents should have an affect no greater than 1,1,1-trichloroethane on the rubber material present in the motors such as silicone but this should be individually examined prior to use.

Therefore it is recommended that hydrocarbon blends such as Evolve CH10 be examined initially, and if this is not effective at removing the soil then either the terpene blend Glidsafe UTS-4B or Evolve CH12 blends be tried. All solvents should initially be tested for materials compatibility with the specific grades of materials present prior to full scale use of a solvent.

9. Conclusion and Recommendations

None of the solvents tested are without some disadvantage. Either they still contribute significantly to ozone depletion, are too volatile, too flammable, too toxic, have high residues, take too long to dry or they are aggressive to some materials. For each application it is therefore necessary to determine what are the important

considerations that cannot be compromised and to balance the other ones to achieve the best result.

For cleaning baluns it is recommended that perchloroethylene be used if appropriate safety equipment and procedures can be put in place, otherwise Evolve CH10 or Glidsafe UTS-4B could be considered. For cleaning electric motors the amount of residue is not as critical and Evolve CH10 should be tried first since it is expected to be the least aggressive on materials. If this is not effective, Evolve CH12, Glidsafe UTS-4B or similar solvent could be considered after checking for any adverse effects on the polymeric materials present in the motor, especially rubbers and varnishes.

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ABSTRACT

The discovery of the hole in the ozone layer over Antarctica and the global drop in stratospheric ozone has prompted the banning of CFC-113 and 1,1,1-trichloroethane, which are common solvents used for degreasing and cleaning electronic components. The Royal Australian Navy uses these substances in a variety of applications and has initiated a replacement program. Two applications, cleaning baluns after lightning strikes and cleaning electrical motors have been selected as priorities under this program. This paper assesses the suitability of various solvents for these applications.

Potential Replacements for Solvents That Are Ozone
Depleting Substances

Lyn E. Fletcher

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