REVISED DRAFT
Hydrocarbon Refrigerants in Australian Cars

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Abstract

Australians need car air conditioning and the hydrocarbon (HC) refrigerant 290/600a avoids stratospheric ozone depletion and a typical 15% increase in TEWI from R134a leakage and service emissions. From nothing in 1994, Australian HC refrigerant sales have increased to about 5% of their available market in 1998 because costs were kept less than half competing refrigerants. Batches of HC replacement for R12, 22 and 502 were manufactured from natural butane and propane by pumping ethane rich vapour off the propane before mixing.

1 Thermal Environment

The Australian islands lie in the Pacific and Indian Oceans with a human population over eighteen million, over ten million motor cars and over six million car air conditioners. The largest and most populous island has an area about the same as contiguous USA but lies across the Tropic of Capricorn. Sydney, Australia, has four million population and outside design temperatures range from 4 to 32°C similar to Los Angeles CA. Other major Australian cities are less temperate and elsewhere on the mainland summer design temperatures range up to 50°C.

Passenger cabins of modern motor cars have insulation to reduce road noise, little thermal inertia and about 3 m^2 of glazing exposed to direct and indirect solar radiation. With windows and doors closed cabins are well sealed except for two air vents at the same elevation and with ventilation fan off and low wind conditions infiltration is less than 1 L/s regardless of the position of any fresh air damper (Maclaine-cross 1997). Cabin temperatures may then rise over 20 K above ambient. The ventilation fan and forward motion may limit this to 10 K but additional measures to avoid heat stress and driver errors are necessary in Australia. The traditional solution is drive with all windows open. The increase in drag coefficient is only about 0.1 but at 72 km/hr this adds 1.4 kW to the crankshaft output required of the engine.

An air conditioner with car windows closed typically loads the crankshaft less than 1 kW at 72 km/hr. In tropical Australia, high speed driving is common and design temperatures are likely throughout the year so air conditioners frequently reduce overall fuel consumption. The Australian Design Rules require the heater/demister components of an air conditioner. The cooling components of an air conditioner add about 3% to vehicle mass which increases fuel consumption for acceleration and rolling resistance by about 2%. In temperate city driving, air conditioners increase fuel consumption.

2 Hydrocarbons Properties

The saturated HCs ethane, propane, isobutane, normal butane, isopentane and normal pentane occur naturally in massive quantities in petroleum gases. The corresponding refrigerant numbers are 170, 290, 600a, 600, 601a and 601 respectively. The Australian market for them as fuels, propellants and solvents is about two million Mg/year. In bulk commercial grades they are cheaper than any other refrigerant except air and water.

The vapour pressure range of HC refrigerants matches that of popular fluorocarbon refrigerants but their significantly lower molecular mass gives them superior transport properties. They are non-hygroscopic and miscible with popular natural and synthetic lubricants. They are good electrical insulators and are compatible with plastic insulation and sealants used in refrigeration systems. They are non-corrosive and chemically stable at temperatures far higher than occur in refrigeration equipment.

HC refrigerants have low toxicity. They are flammable but the necessary precautions and consequences have been considered elsewhere and long ago (Maclaine-cross 1996, 1997). In 1992, HCs were revived as commercial refrigerants because they do not deplete stratospheric ozone and have negligible global warming.

In the atmosphere, HC refrigerants are highly reactive and within days are converted to carbon dioxide, water vapour and methane. Methane itself becomes carbon dioxide and water vapour after several years. Averaged over the troposphere, refrigerant HCs reduce ozone concentration although other HCs increase tropospheric ozone (Johnson and Derwent 1996). A simple calculation of global warming potential (GWP) is to neglect the effects of methane and ozone and assume carbon dioxide and water vapour form immediately after emission. Johnson and Derwent (1996) have calculated a 100 year global warming potential for a step increase in emissions (SGWP) which includes the indirect warming effects of both methane and ozone. The two are compared below:

Chemical name	ethane	propane	butanes	pentanes
Refrigerant	170	290	600(a)	601(a)
Simple GWP	2.93	2.99	3.03	3.05
SGWP (J. & D. 1996)	4.3	1.1	2.7	3.0

In the SGWP above, the reduction in indirect global warming from ozone partially cancels the increase from methane. We recommend 3 as a representative GWP for HC refrigerants. The SGWP are higher than traditional impulse GWPs but more realistic (Johnson and Derwent 1996). We recommend that GWPs for HC refrigerants be calculated from their mass fraction composition and the SGWP above.

3 Emission Reductions

Fluorocarbon emissions can be calculated from government or industry import records since Australia does not manufacture these chemicals. Import statistics for fluorocarbons are available from the Australian Bureau of Statistics only after payment of substantial charges. Published estimates of fluorocarbon emissions by mobile air conditioning are based on assumptions about the vehicle population, leakage and service emission rates which may not be representative. These estimates are 1765 Mg in 1986 and 2037 Mg in 1996. Estimated total fluorocarbon gas consumption has however declined from 17333 Mg in 1986 to 4153 Mg in 1996.

Fluorocarbon gases have almost disappeared from the large foam, solvent and propellant markets. The mix of fluorocarbons has changed. Imported R22, 123 and 134a have replaced locally manufactured R11, 12 and 22. Australia has massively reduced its ozone depleting emissions. The reduction in global warming emissions from refrigerants could however be much greater if fluorocarbons were replaced entirely in car air conditioners.

Measurements at Cape Grim, Tasmania show that global atmospheric concentrations of ozone depleting refrigerants are still rising and their fluorocarbon replacements are growing exponentially (Fraser 1997, 1998). A major contributor to this are countries not covered by Montreal Protocol restrictions. A 1998 bulk price for R12 in India was only 1.5 US $/kg and Indian R12 marketed in Indonesia was 3 US $/kg. In 1998 bulk R134a was available in Indonesia for 4 US $/kg.

Aisbett and Pham (1998) have projected the environmental impacts of car air conditioners for thirteen Asian countries with half the world's population. They have considered three scenarios for replacement of CFCs from 1985 to 2020. Their scenario two is replacement with HFCs and scenario four replacement with HCs. By 2020, the HFC scenario gives a total environmental warming impact 286 million Mg/year greater than the HC scenario. More importantly the cost of HFCs is 3.5 billion US $/year greater than HCs mostly in hard currencies.

4 Car Refrigerant Emissions

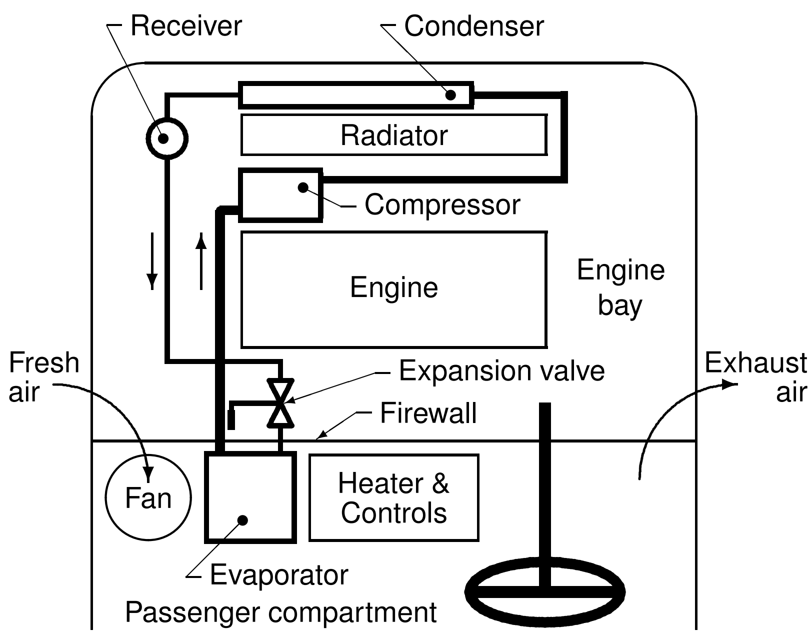

Figure 1: Schematic of car air conditioner with liquid line receiver and expansion valve in engine bay (right hand drive).

Car air-conditioners cool with a simple reversed Rankine cycle. Major components, refrigerant and air flows are shown schematically in Figure 1. A compressor bolted to the engine has an open drive for efficient power transfer and flexible refrigerant hoses to limit vibration stresses. Permanent Schrader valves are provided in both low and high pressure lines, so refrigerant may be added, air released and pressure gauges attached.

A domestic refrigeration appliance leaks less than 0.001 L/year of liquid because the drive motor is sealed inside the circuit. Open drives, hoses and Schrader valves on cars together leak 0.1–1.0 L/year of liquid. Repairs are however usual if leaks exceed 0.4 L/year. A reservoir containing up to 0.5 L of extra refrigerant liquid allows continued operation for several years between recharging. A full charge is 0.4–1.0 L liquid.

Releases of refrigerant to atmosphere must be added to leakage to calculate average emission rate. Releases may occuring during manufacture, handling, shipping or air conditioner servicing. Releases during manufacture, handling and shipping may not be small but depend only slightly on the containers used and hence service procedures. Service releases are frequently many times annual leakage. They depend on charging procedure, skill and effort. The main charging procedures are ‘top-up’, ‘recovery’ and ‘regas’. Service release from the ‘top-up’ charging procedure are less than 10 mL liquid per service with low skill and effort. Releases from ‘recovery’ procedures can be less than 10 mL liquid per service with high skill and effort. Releases from ‘regas’ procedures may be more than the total refrigerant charge or over 1 L liquid per service if little leakage has occurred since the last regas.

Many small garages and do-it-yourself technicians in North America favour the ‘top-up’ charging procedure. Even with traditional tools and fittings the procedure is simpler and easier than changing the engine oil and filter. When the low pressure cut-out switch prevents the air conditioner operating about 0.3 L liquid refrigerant is added from a premeasured can. The cans may contain 2% by mass lubricant to make up lubricant lost with leaking refrigerant. Annual total emissions are then just above the leakage rate and are typically half those for regassing. An 0.3 L steel can is readily recycled by modern recycling machinery even if it is accidently full. The container mass is about 80 g less than 50% of HC refrigerant mass compared with about 10 kg or 110% for 20 L reusable cylinders saving substantially on transport energy and pollution. However top-up is rarely used in Australia.

For CFCs and in one Australian state for HCFCs, regulations require ‘recovery’ procedures. These procedures use a vacuum pump for typically 30 minutes to remove almost all refrigerant from the system and store it in a cylinder before recharging. The procedure removes a large part of the lubricating oil and contaminants from the system with the refrigerant. The commercial value of contaminated refrigerant is negligible. The most profitable legal use of contaminated refrigerant is to distill most of it for reuse on site and then return the highly contaminated

Table 1: Carbon dioxide equivalents of car air conditioner emissions.

Refrigerant	12	134a	290/600a
ODP (R11 ODP=1)	0.82	0.00	0.00
GWP (100 year)	8500	1300	3
Liquid density (kg/m^3)	1300	1206	523
Refrigerant emission (g/year)	520	482	209
CO_2 equivalent (kg/year)	4420	627	0.6
Ratio to fuel CO_2 (%)	102.8	14.6	0.0

cylinder to the supplier for cleaning. Such distillation and cleaning are expensive, labour and energy intensive processes. Annual emissions from recovery lie between those for top-up and regas. Enforcement in Australia increased costs but was not vigourous enough to reduce recovery emissions significantly below regas. The most effective enforcement measure, a substantial tax on new refrigerant was never used.

Car air conditioners have no level gauge to indicate the refrigerant remaining before service. The usual ‘regas’ procedure vents any remaining charge to atmosphere and weighs out a complete new charge. It is legal for R134a throughout Australia. The total emissions are then the slow leakage between regassing and the release of the remaining charge during regas. By conservation of mass these must equal the charge added at the previous regas. Annual total emissions with regassing are thus refrigerant charge divided by the regas interval in years. With regassing the leakage rate has no effected on the average total emission rate.

With regassing comparison of refrigerant environmental impacts for any car air conditioner is easy. Table 1 compares refrigerants for a car using 2000 L/year of fuel, having an 0.8 L liquid refrigerant charge and regassed every second year. These assumptions are typical but representative values are not known for Australia (Section 3). One litre of petrol burns to 2.15 kg of carbon dioxide so the fuel consumption emits 4300 kg/year of carbon dioxide. Annual total refrigerant liquid emissions are $0.8/2 = 0.40$ L/year for each refrigerant in Table 1.

5 Photochemical smog and ozone

The exhaust from engines using HC fuel contains small amounts of carbon monoxide, HCs and oxides of nitrogen. HCs may also evaporate from the fuel tank and supply system. If sunlight falls on air with high concentrations of nitrogen dioxide and HCs, ozone is formed as a byproduct of HC oxidation and nitration (Carter

1994). These reactions occur with both light and heavy HCs and halons like HFCs and HCFCs (Derwent *et al.* 1996). Ozone concentrations drop after sunset and as HCs become oxides or nitrogen oxides become nitrates.

For heavy halon and HC reactants like liquid fuels, the vapour pressure of the products may be low enough for condensation to a fog, called smog. Refrigerant halons and HCs are light so their product vapour pressure is too high to condense to smog (Avallone and Baumeister 1986).

Johnson and Derwent (1996) found that emissions of refrigerant HCs reduce tropospheric ozone as a whole. When wind is present or sunshine or traffic absent HC refrigerant emissions reduce ozone in cities. For air pollution episodes, emitted refrigerant HCs create less ozone than exhaust HCs (Carter 1994, Derwent *et al.* 1996). For HC refrigerant in car air conditioners to increase ozone during such episodes, their rate of emission would need to be greater than the saving in HC exhaust emissions.

Few HC car air conditioners leak more than 200 g/year or 6.34μg/s. If averaging 20 km/hour in traffic this is 1.14 mg/km. Maclaine-cross and Leonardi (1996) found an average fuel saving on converting from R12 to HC refrigerant was about 1.3%. Exhaust HC emissions of 100 mg/km are well inside current Australian standards for new cars (FORS 1995). We expect the saving in exhaust HC emissions to be proportional to fuel savings or 1.3 mg/km. The net saving in HC emissions is 0.16 mg/km. Typically the reduction in HC emissions from converting to HC refrigerant is much greater.

HC refrigerants in car air conditioners reduce photochemical smog and ozone. These environmental advantages are small compared to reduced global warming and stratospheric ozone depletion.

6 Marketing strategy

Kuijpers *et al.* (1988) revived the idea of using HCs and their mixtures as refrigerants. In 1992, HC mixtures were used in the Foron refrigerator sold in Europe and in 0.3 L cans for car air conditioners sold from Idaho USA by OZ Technology.

In 1993, the fluorocarbon industry staged spectacular stunts for television in Florida, California and elsewhere. A passenger cabin was filled with about 500 g of OZ-12 which was ignited blowing some windows off. OZ Technology replied with a stunt video of their own, demolishing a cabin with R134a.

The US Bill of Rights guarantees citizens and corporations equal treatment before the law. Among other things this requires that if A breaks a particular law before B, A must be prosecuted before B. R134a was put into car air conditioners in 1990, two years before OZ-12. Prosecutions of OZ Technology, initiated under US state laws prohibiting flammable refrigerants, collapsed because the OZ video convinces anyone that R134a is legally flammable. No State Attorney-General would prosecute R134a suppliers first just so they could prosecute OZ Technology. In 1997, Idaho amended its law to permit both HCs and R134a. The relevance for HC marketing strategy is keep engineering, production and distribution costs low so you can afford good lawyers and videos.

Late in 1993, Australia's fluorocarbon lobbyists began distributing the exploding HC car videos and resulting press reports to journalists and key Australian government officials. By early 1995, they had State governments competing to prohibit HC refrigerants first but no HC refrigerant sales to prohibit. Queensland was first to make an exploding car video. On the 26th October 1995, New South Wales released their own exploding car video and became the first Australian state to restrict HC refrigerants. The Victorian government, including the EPA, Fair Trading, Fire Brigade, Vicroads and Workcover, requested scientific evidence to support proposed regulations however none was supplied. HCs are still severely restricted in NSW and Queensland over half the Australian refrigerant market.

In early 1994, Calor introduced the CARE range of HC refrigerants in Europe. In November 1994, Elgas decided to produce the CARE range in Australia. In mid-1995, both OZ and CARE ranges were available in Australia at similar prices to fluorocarbon refrigerants.

In October 1994, two air conditioning technicians read about European and US developments. They decided HC refrigerants were safer, more efficient and should be half the price of fluorocarbon refrigerants. In December 1994, they made their own samples and distributed them to trade colleagues. By July 1995, they had formed Esanty and had their product on the market at half the price.

Esanty knew refrigerant is a large fraction of the costs of servicing car air conditioners. This service trade has personal customer contact and its business is more local and loyal than that of manufacturers. Its business is less easily damaged by whisper campaigns and hostile press releases. Esanty consulted all sections of the industry which agreed that acceptance by the service trade was a prerequisite to vehicle manufacturer acceptance.

The trade wanted a replacement for R12 which was as much like R12 as possible. It wanted the same cylinders, fittings, tools and lubricants. Traditionally they used 20 L cylinders. When it gets hot people want their air conditioners fixed, the trade wants full cylinders and distributors order pallets. In Victoria 80% of sales occur in Spring and Summer. If distributors are out of stock of one refrigerant, the trade buys another so success required short supply lines and simple high speed manufacturing methods.

7 Manufacture

Total Australian sales of HC refrigerant are small. Combining data from the three main manufacturers and allowing for imports and a number of smaller manufacturers our estimates are:

Calendar year	1995	1996	1997	1998
Domestic sales (Mg)	3	11	18	29

Up to 1998 almost all sales were in Victoria to the car air conditioner market. The 1998 sales represent only 1.5% of the total Australian refrigerant market but about 5% of the market legally available to HCs. Exports sales for Australian suppliers are much smaller than domestic sales. Only batch production is economic for such small volumes.

The basic cost of mass producing HC refrigerants is less than 1% of fluorocarbons (Aisbett and Pham 1998). Table 2 is the specification for HC refrigerant used in Australian car air conditioners. Australia produces over three million Mg of commercial propane and butane a year and most of this is naturally occurring. Naturally occurring sources have a very low content of unsaturated HCs and other impurities. Commercial propane contains 2–6% ethane by mass. In 1995, there was no commercial secondary separation and purification of isobutane in Australia. Supplies with odorant added are cheaper because of reduced road tanker handling costs. In the workshop, odorant is also a useful safety precaution. Prices vary but are usually below 0.30 US $/kg delivered by road tanker.

High purity HCs are about 3 US $/kg into store. Small quantities travel as general container cargo so delivery usually takes about two months but sometimes months more. Substantial capital is imprisoned on the high seas.

Temperature glide is the difference between the dew and bubble point temperatures at a given pressure. The specifications in Table 2 important to performance are the boiling or bubble point pressure and the temperature glide. Pure R290/600a [50/50] has a glide about 8 K. Lower glide gives lower boiling pressure at which evaporator superheat can be achieved when replacing R12 in car air conditioners. Ethane greatly increases glide.

R170/290 [6/94] is a good replacement for the CFC R502 used in commercial refrigeration. Commercial propane usually contains more than 2% ethane so it cannot be mixed with commercial butane to replace R12 and less than 6% ethane so it cannot replace R502. The ratio K of ethane mass fraction in equilibrium vapour to that in liquid commercial propane is about three (Ely and Huber 1992). A compressor with 1 L/s displacement can pump over 1 Mg/day of this vapour through a 6 kW condenser coil submerged in the propane liquid. A trap on the coil exit should prevent vapour entering the condensate tank (Figure 2). For 2.0–2.5% ethane in the commercial propane, the condensate may be repumped to get

Table 2: HC refrigerant quality specification for car air conditioners (updated from IAHRA 1996).

Component	Unit	Specification Tolerances
Saturated HCs	% mass	> 99.5
Ethane	% mass	< 2.0
Propane	% mass	–
Isobutane	% mass	–
Normal butane	% mass	–
Pentanes	% mass	< 0.5
Normal hexane	mg/kg	< 100
Aromatics	mg/kg	< 10
Unsaturated HCs	% mass	< 0.05
Water	mg/kg	< 10
Ethyl mercaptan	mg/kg	25
Other sulphur compounds	mg/kg	< 5
15°C bubble point pressure	kPa	480–570
Temperature glide	kelvin	< 12
Lubricant additive	% mass	< 2

a product with 6% ethane. Commercial propane feed with 3% or less ethane is desirable to avoid producing more R170/290 than can be sold.

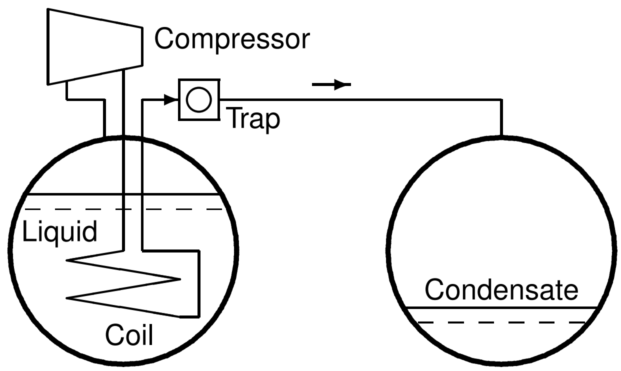

Figure 2: Schematic of batch pumping of ethane rich vapour off propane liquid to lower ethane mass fraction in the liquid.

Conservation of ethane and propane mass in the liquid gives the final mass fraction of ethane in the liquid x_f as a function of the initial mass fraction x_i as

$$x_f = x_i \left(\frac{m_f}{m_i} \right)^{K-1} \tag{1}$$

where m_i is the initial, m_f the final liquid mass and K is assumed constant. Conservation of ethane and propane mass overall gives the average ethane mass fraction in the condensate and vapour phases x_c as

$$x_c = \frac{x_i m_i - x_f m_f}{m_i - m_f} \tag{2}$$

If the volume of the vapour above the liquid is less than half the liquid tank volume when pumping finishes, x_c is close to the ethane mass fraction in the condensate tank. As an example consider $K = 3$, $x_i = 3.00\%$ and $m_f/m_i = 0.6181$ from Equation 1, $x_f = 1.146\%$, and from Equation 2, $x_c = 6.00\%$.

HC replacements for R12, 22 and 502 are manufactured in Australia. The compressor, coil and trap above are the additional major equipment items necessary to use commercial Australian HCs instead of highly refined imported supplies. This method has resulted in manufacturing costs less than half that of landing fluorocarbons even though fluorocarbons have fifty times the sales volume in Australia. These Australian HC refrigerants cost less than 1 US $/kg to produce and are still significantly cheaper than HFCs and even Indian CFCs after shipping to South-East Asia.

In 1998, commercial secondary separation of Australian butane commenced. R600a is energy-efficient in small appliances (Maclaine-cross and Leonardi 1997) and their manufacturers prefer pure isobutane. Pure isobutane has now replaced commercial butane in Australian R290/600a replacements for R12.

Safety precautions in manufacturing Australian HC refrigerants are the same as in the LPG fuel industry (Maclaine-cross 1996). Documentation, sites and equipment are well established and much cheaper than other chemical plant for the same level of safety.

8 Distribution

HCs have no inherent cost advantages in distribution. Freight charges depend on volume not mass. Almost all traditional refrigerant distributors in Australia refuse to sell HCs. Traditional refrigerant suppliers own the cylinders and rent them to their customers.

The fluorocarbon industry invented the 0.3 L can technology decades ago. They never promoted it perhaps because reduced emissions from top-up mean reduced sales. OZ Technology of Idaho improved this technology with instructions, tags and added lubricant. Contract propellant canners using existing equipment in Australia can ship pallet loads for about 8 US $/kg HC refrigerant.

Australian HC suppliers chose the 20 L cylinder technology for acceptance by the existing market. Their refrigerant distribution improvements include:—

- Distribution with other car parts and supplies through traditional automotive distributors.
- Encouraged new refrigeration distributors specializing in environmentally acceptable technology.
- Encouraged LPG cylinder manufacturers to use their higher volume to make refrigerant cylinders at lower cost.
- A US $120 deposit on each 20 L cylinder instead of rental. The accounting systems of car parts distributors handle deposits but not always rental.
- Cylinders are packed in a cardboard carton which is usually removed and replaced at the cleaning and filling plant. This reduces handling damage to valves, paint and labels.
- Instructions are packed in the crate with each cylinder.
- Double valves to prevent valve leaks in the distribution chain.

- Ethyl mercaptan odorant warns of leaking cylinders.
- Safety improvements and advice and information provided to customers reduce the cost of public liability insurance.

Even with these improvements pallet loads of full cylinders ship for 11 US $/kg HC refrigerant.

Cans are very suitable for top-up charging of car air-conditioners using half the refrigerant with half the emissions of the regas method typically used with cylinders (Section 4). With cans a trader typically uses half the mass of refrigerant servicing the same number of vehicles as with cylinders. Since labour is also less with cans a trader could pay twice as much per kilogram for refrigerant in cans and still profit. The price per kilogram for pallet loads of HC refrigerant in cans is actually less than for cylinders. The higher margins available to distributors have rapid expanded HC can sales in North America since 1992. No refrigerant canning currently occurs in Australia.

9 Conclusion

Australians need car air conditioning but total refrigerant emissions from leakage and service releases are typically 0.4 L/year of liquid for regas. If the refrigerant is HFC–134a this adds about 15% to the car's total global warming emissions. For Asia the refrigerant is typically CFC–12 adding about 102% to emissions. HC refrigerants can reduce this warming to zero and slightly reduce tropospheric ozone.

In 1992, HC refrigerants were successfully reintroduced in Europe and the USA. In 1995, when HCs arrived in Australia, fluorocarbon lobbyists cajoled two out of eight regional governments into severely restricting them. Australian HC suppliers survived in only half the potential market by innovations in manufacture and distribution to keep their prices half those of fluorocarbon suppliers.

The most important innovation was to use commercial butane and propane as feedstocks which are naturally abundant with high purity in Australia. Initially the only secondary processing apart from mixing was to 'pump off' ethane from the commercial propane. This gave HC replacements for R12, 22 and 502 from the same plant.

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