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International Research into Chlorofluorocarbon (CFC) Alternatives

T. J. Marseille
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May 1992

Prepared for the U.S. Department of Energy
International Research Monitoring Program
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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MASTER

EXECUTIVE SUMMARY

Because of an international awareness of the potentially destructive impact that chloroflourocarbons (CFCs) and other halocarbons have on stratospheric ozone, industrial nations have agreed to a timetable for the phaseout of CFCs, halons, and related components. This timetable, included as part of the international treaty called the Montreal Protocol (Marseille and Baechler 1990), has acted as a driving force in instigating worldwide research activities into CFC alternatives. To explore the scope and direction of these activities, Pacific Northwest Laboratory conducted a survey for the U.S. Department of Energy on international research into alternatives for CFCs.[a]

Selected researchers from 21 countries were queried through questionnaires about their current and planned research activities. The results of the survey show that the majority of research being conducted by the respondents is devoted to investigating the hydrogenated fluorocarbon HFC-134a as a replacement for CFC-12 in refrigeration applications. The main issue with this alternative is identifying compatible lubricants that do not reduce its effectiveness.

Although researchers in different countries are often investigating similar alternatives, they appear to be at different stages of research. For example, some researchers are at more advanced development stages, conducting laboratory and field tests of alternatives in end-use applications, while others are still conducting more basic research, such as analyzing the thermal and chemical properties of a specific alternative. Improved international communication could help bridge this discontinuity in research progress. Moreover, the survey results indicate there is a strong interest in learning about the status and results of the research efforts of other researchers.

(a) This report was prepared through the International Research Monitoring Program (IRM), which was established in 1985 to monitor and evaluate the current status and future directions of international research and development activities of interest to the U.S. Department of Energy's Conservation and Renewable Energy Program (DOE/CE). The IRM Program is managed by the DOE/CE Office of Technical Affairs with technical assistance from Pacific Northwest Laboratory.

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1.0 INTRODUCTION

Chlorofluorocarbons (CFCs) are chemically stable members of the halocarbon family that are derived from common hydrocarbons such as methane, ethane, and propane. The halocarbons also include hydrogenated chloroflourocarbons (HCFCs), hydrogenated fluorocarbons (HFCs), brominated hydrocarbons (or halons), as well as miscellaneous chlorinated hydrocarbons such as carbon tetrachloride and methyl chloroform. Of all the halocarbons, CFCs are of greatest concern with regard to stratospheric ozone damage.

About 30% of CFCs produced worldwide are used as the refrigerant working fluids in vapor-compression cycle air conditioning and refrigeration systems and as blowing agents (28%) in the production of insulating foams and packaging. They are also used for aerosols (19%), as solvents for electronics cleaning (19%), and for other miscellaneous uses (4%). Primary industries that use CFCs include commercial and residential construction, food processing, transportation, electronics, and plastics. Altogether, worldwide annual production of CFCs reached approximately 2 billion pounds in 1986 in non-centrally planned economies (Moore 1989).

CFCs have life expectancies on the order of 100 years. It is theorized that over time individual CFC molecules that have been released on the earth's surface diffuse into the stratosphere where they will break down under ultraviolet (UV) bombardment from the sun (Rowland and Molina 1975). Once broken down, the halogen chlorine, found in all CFCs, disrupts normal stratospheric chemistry, causing ozone destruction. It has been estimated that every free atom of chlorine can participate in the destruction of 100,000 ozone molecules (Rowland and Molina 1975). This destruction is environmentally significant because the "ozone-layer" in the stratosphere normally acts as a protective shield to the earth, effectively blocking the sun's harmful UV rays from reaching the surface. The U.S. Environmental Protection Agency (EPA) has predicted that every 1% decrease in ozone will result in a 1% to 2% increase in the incidence of melanoma skin cancer (EPA 1987). Other expected effects of ozone depletion include crop damage, increased incidence of cataracts, increased formation of tropospherical ozone (smog), and increased degradation of manmade materials such as polymers (EPA 1987). The term *ozone depletion potential* (ODP) has been developed to provide a relative measure of the detrimental effect that different chemicals have on the ozone layer.

The second threat caused by the release of CFCs into the atmosphere is global warming, also known as the greenhouse effect. Concentrations of various gases found in the atmosphere, known collectively as greenhouse gases, are increasing because of human activities. Among these gases are CFCs and carbon dioxide (CO_2). Because of these larger concentrations, a larger portion of the long-wave radiation emitted from the earth's surface that previously escaped into space is absorbed by these gases, producing additional heat that warms the atmosphere. The term *global warming potential* (GWP) have been developed as a relative measure of the potential of various greenhouse gases to absorb long-wave radiation. The GWP of CFCs is as much as 10,000 times as large as that of CO_2, which is by far the most prevalent greenhouse gas. Thus, though much smaller concentrations of CFCs than CO_2 are present in the atmosphere, it is estimated that CFCs contribute approximately 10% to 15% to the greenhouse effect (WMO 1985). The ODPs, GWPs, and atmospheric life expectancies of each of the major halocarbons are summarized in Table 1.1.

In CFCs, all of the hydrogen atoms surrounding the carbon atoms in the original hydrocarbons are replaced by either fluorine or chlorine, which are two halogen elements. The HCFCs are similar to CFCs except that some of the original hydrogen atoms remain. These compounds generally contain less chlorine and break down sooner in the atmosphere than CFCs. Thus, HCFCs have lower ODPs, which means that they have less of an impact on the ozone layer. Their shorter atmospheric lifetimes also result in lower GWPs, which reduces global warming as well. In addition, many HCFCs have thermodynamic and physical properties similar to those of CFCs. These lower ODP and GWP CFCs, at least in the near term, appear to be attractive potential candidates for replacing CFCs in many applications.

The HFCs are fluorine compounds that are similar to HCFCs and CFCs except they contain no chlorine. Consequently, they have ODPs of zero. They also have shorter atmospheric lifetimes than CFCs and, thus, lower GWPs.

TABLE 1.1. Environmental Effects of CFCs and Possible Alternatives

Fluid Number Name	Formula	ODP[a]	GWP[b] 100 year	GWP[b] 500 year	Atmospheric Life(c) (year)
HFC-23	CHF_3	0		12,000[d]	310[d]
HFC-32	CH_2F_2	0		220[d]	6[d]
HFC-125	CF_3CHF_2	0	2,500	860	28
HFC-143a	CF_3CH_3	0	2,900	1,000	41
Propane	$CH_3CH_2CH_3$	0	3[e]	3[e]	<1
HCFC-22	$CHClF_2$	0.05	1,500	510	15
Ammonia	NH_3	0			<1
HFC-134a	CF_3CH_2F	0	1,200	420	16
HFC-152a	CHF_2CH_3	0	140	47	2
HFC-134	CHF_2CHF_2	0			12[d]
HFC-227ea	CF_3CHFCF_3	0			
HCFC-124	$CHClFCF_3$	0.02	430	150	7
Isobutane	$CH(CH_3)_3$	0	3[e]	3[e]	<1
HCFC-142b	$CClF_2CH_3$	0.06	1,600	540	19
n-butane	$CH_3(CH_2)_2CH_3$	0	3[e]	3[e]	<1
E-134	CHF_2OCHF_2	0			<12
HCC-160	CH_2ClCH_3	0.02	3	3	<1
HCFC-123	$CHCl_2CF_3$	0.02	85	29	2
E-245	$CF_3CH_2OCHF_2$	0			
HCFC-141b	CCl_2FCH_3	0.10	440	150	8
Carbon Dioxide	CO_2	0	1	1	120
CFC-13	$CClF_3$				400
CFC-115	$CClF_2CF_3$	0.35	6,900	7,400	400
CFC-12	CCl_2F_2	0.9	7,300	4,500	130
CFC-114	$CClF_2CClF_2$	0.7	6,900	5,500	200
CFC-11	CCl_3F	1	3,500	1,500	60
CFC-113	$CClF_2CCl_2F$	0.8	4,200	2,100	90

(a) ODP values are relative to R11 and are averages of the 2-dimensional model results presented in WMO Report No. 20, Vol. 11 (1989).

(b) Global warming potentials are from the IPCC Scientific Assessment (1990), except as noted; values are relative to CO_2 and are given for 100 and 500 year integration time horizons.

(c) Atmospheric lifetimes are from WMO Report No. 20, Vol II (1989), except as noted.

(d) Estimates based on limited data, GWP is steady-state value (McFarland, personal communication).

(e) GWP of the hydrocarbons is due almost entirely to GWP of the CO_2 resulting from decomposition.

Therefore, the use of these substances in applications that currently employ CFCs is environmentally highly desirable.

Halons differ from CFCs in that they include bromine, the heaviest halogen element. Bromine has a higher potential for ozone damage that chlorine; thus, halons are usually included in environmental discussions about CFCs for completeness. Halons are used primarily as a fire-extinguishing agent in total flooding systems for critical civilian electronics installations and military applications. Researchers queried for this study were exclusively looking at alternatives for CFCs; therefore, halons will not be discussed further in this report.

In response to the recognized threat CFCs pose to the ozone layer, over 60 nations, including major world economies that produce and use CFCs, participated in drafting an international treaty known as the Montreal Protocol on Substances that Deplete the Ozone Layer. The treaty originally committed signatory nations to reduce the production of CFCs, halon gases, carbon tetrachloride, and methyl chloroform by 50% by the year 1998.

More recent scientific evidence has indicated an even higher ozone-depleting potential for chlorine and bromine compounds than originally suspected, prompting further international research. A March 1989 meeting of the European Economic Community (EEC) called for a complete CFC phaseout in production by 2000 and a complete phaseout in production of HCFCs by 2005. The signatory parties to the Montreal Protocol met in Helsinki, Finland, in May 1989 and approved a declaration that stated a similar intent to phaseout consumption of CFCs by the year 2000 (assuming acceptable alternatives are available) and to refrain from using other halons except for essential applications. In London in June 1990 the Second Meeting of the Parties to the Montreal Protocol on Substances that Deplete the Ozone Layer resulted in the adoption of these more stringent amendments, summarized in Table 1.2. In addition, it was resolved at the London meeting that guidelines would be applied to facilitate the use of HCFCs in place of CFCs where necessary. These guidelines would include the regular review of alternative products to HCFCs, with a view to replace HCFCs no later than 2040 and, if possible, no later than 2020.

TABLE 1.2. Phaseout Schedules Adopted at the June 1990 United Nations Environmental Program (UNEP) Meeting in London

Compound	Goal
CFCs	Production phaseout by year 2000 if environmentally acceptable alternatives are available
Halons	Phaseout by 2000, unless satisfactory substitutes are identified for essential applications
Carbon tetrachloride	Production phaseout by year 2000 if environmentally acceptable alternatives are available
Methyl chloroform	Production phaseout by year 2005.

Clearly, the progress toward the development of alternative compounds will have a significant impact on the effectiveness of the treaty. To assess this progress, Pacific Northwest Laboratory (PNL)[a] conducted a study for the U.S. Department of Energy (DOE) International Research Monitoring Program to obtain a global perspective on and document foreign research activities into CFC alternative chemical compounds.

Section 2.0 of the report describes the methodology used to survey scientists. The results of the survey are then described in Section 3.0, with conclusions presented in Section 4.0. A copy of the questionnaire is contained in Appendix A. Appendix B contains the cover letter, and Appendix C contains a list of who was sent the questionnaires.

(a) Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

2.0 METHODOLOGY

The goal of this study is to document international research activities being conducted to find replacements for CFCs. Although this information is widely available from conference proceedings and other published references, frequently the information is out-of-date because of the length of time from submittal to publication. Therefore, a mail questionnaire was developed to provide the most current information on international research activities and to gather more detailed information than normally provided in research articles.

The questionnaire asked researchers to provide information about their specific research projects regarding CFC replacements, the results of the research to-date, problems encountered and how these were overcome, current and projected funding levels for their research as well as their sources for funding, and potential research projects in the near future. A copy of the questionnaire is in Appendix A.

The list of international researchers was generated from a literature review of all recently published international research on CFC replacements. Some of the sources include the June 1991 issue of the International Energy Agency (IEA) Heat Pump Centre Newsletter, the proceedings from the 1990 International Conference on CFC and Halon Alternatives, and the 1990 U.S. National Committee for the International Institute of Refrigeration (USNC/IIR) Purdue Conference.

The questionnaire was sent to all international researchers that published within the last 5 years. In addition to the literature review, contacts were made with U.S. researchers and staff at DOE to identify other international researchers. Due to sensitivity issues, several countries who are known to be conducting research into CFC replacements (e.g., USSR) were not sent questionnaires; therefore, their current research is not included in the report.

The questionnaire and a cover letter (see Appendix B) explaining the purpose of the questionnaire were sent to 131 individuals in 21 countries. A list of those sent the questionnaire is contained in Appendix C. The questionnaire was sent during March 1991. As of June 1991, 81 questionnaires (62%) were returned. A listing of the number of researchers who returned the questionnaire by country is provided in Table 2.1.

Although an attempt was made to identify all international researchers studying replacements to CFCs, unfortunately some will have been overlooked. The results of this study should be viewed as providing an overview of current international research activities on CFC replacements; not a complete documentation of all research efforts.

TABLE 2.1. Number of Researchers Contacted and the Number that Responded by Country

Country	Number of Contacts	Number of Respondents
Australia	2	2
Austria	2	2
Brazil	5	3
Canada	17	6
China	3	1
Denmark	4	4
France	3	3
Germany	14	10
India	4	3
Italy	2	1
Japan	35	23
Kenya	1	1
Korea	1	1
Mexico	3	1
New Zealand	1	1
Norway	6	4
Sweden	5	3
Switzerland	3	2
Taiwan	1	1
The Netherlands	3	2
United Kingdom	15	8

3.0 RESULTS OF THE SURVEY

This section describes the results of the survey. Some of the answers provided on the questionnaire were difficult to decipher, primarily due to the language barrier. Also, some of the respondents offered little detail in their answers.

3.1 LENGTH OF RESEARCH

Most of the respondents indicated that they began research into CFC-related issues 3-5 years ago. Most also expected to continue research activities for 3 or more years. These results indicate that the CFC issue is recognized as a serious international issue, and that identifying and then developing alternatives are requiring an intensive, ongoing research effort. Table 3.1 contains the respondents' answers to the questions about the length of time of their research.

3.2 IDENTIFICATION OF ALTERNATIVES

Although many CFC alternatives are being studied, various HCFCs, HFCs, and mixtures of the two are by far the most widely researched. The most prevalent alternatives being studied by the respondents were HFC-134a and, to a lesser extent, HCFC-22. Table 3.2 lists (by percent) what CFC alternatives were identified in the surveys as having a high potential for alleviating the

TABLE 3.1. Length of Time that Respondents Have Been Conducting Research (Percent of Respondents)

	How Long Research Has Been Conducted	How Long Research Will Continue
More than 5 years	16%	35%
3 - 5 years	47%	31%
1 - 2 years	17%	11%
Less than 1 year	4%	7%
Missing	16%	16%

TABLE 3.2. Specific Alternative Compounds that Respondents Believe Offer the Most Potential for Alleviating the CFC Problem

Alternative Compound	Respondents
HFC-134a	36%
HCFC-22	9%
HFC-152a	5%
Ammonia	4%
Other	20%
Missing	26%

CFC problem. Specifically, over one-third of the respondents reported the alternative HFC-134a as having the most potential for alleviating the CFC problem.[a] About 9% of the respondents listed HCFC-22 as the most promising alternative, and a few of the respondents mentioned either HFC-152a or ammonia (NH_3).

Most of the respondents from Japan, Germany, and the United Kingdom are analyzing HFC-134a. However, a large number of Japanese and United Kingdom respondents did not identify the specific alternative(s) they were researching. Table 3.3 lists specific CFC alternatives respondents from individual countries chose as providing the most potential for alleviating the CFC problem. Only those countries with three or more responses are listed.

(a) Interestingly, though HCFCs were often cited by researchers as alternatives, these chemicals offer at best only a temporary solution because they still contain some chlorine. Because of this, their eventual production phaseout is also expected by 2020-2030.

TABLE 3.3. Specific Alternative Compounds that Respondents in Each Country Believe Offer the Most Potential for Alleviating the CFC Problem

Country	Percent	Alternative Compound
Japan	48	HFC-134a
	9	HCFC-22
	4	Aqueous Cleaning
	4	HCFC-123
	35	Missing
Germany	50	HFC-134a
	20	HFC-152a
	10	R718
	10	Hydrogen
	10	Missing
United Kingdom	25	HFC-134a
	13	HFC-152a
	13	ISCEON 69S
	13	Ammonia
	38	Missing
Canada	33	HCFC & HFC
	33	HFC-134a
	17	HFC-152a
	17	Missing
Denmark	50	HFC-134a
	50	Missing
Norway	50	HCFC-22
	25	Ammonia
	25	Missing
Brazil	67	HFC-134a
	33	Missing
India	33	Azeotrope
	33	HCFC-22
	33	HFC-134a
Sweden	33	HFC-134a
	33	NARMs
	33	Ammonia

3.3 PURPOSE OF THE RESEARCH

Most of the respondents indicated they are currently investigating the application of one or more CFC alternatives to vapor-compression cycle refrigeration/air-conditioning equipment. Of these respondents, about half are looking at HFC-134a. Over one-third of the respondents are conducting theoretical performance analyses of the vapor-compression cycle. Of these, almost half are studying HFC-134a and a smaller percentage are studying HCFC-22. About one-third of respondents reported analyzing thermodynamic properties of alternatives, and one-third are studying the compatibility of the various alternatives with materials used in air-conditioning and refrigeration equipment. Again HFC-134a is the dominant compound being analyzed for both purposes. Table 3.4 lists the different purposes of the research. Because of the high frequencies of HFC-134a and HCFC-22 on the surveys, additional information is presented here.

TABLE 3.4. Purpose of the Research

Purpose	All Respondents	HFC-134a Respondents	HCFC-22 Respondents
Test Applications	57%	69%	57%
Refrigeration/Air-Conditioning	64%	86%	86%
Insulation/Blowing Agent	10%	3%	14%
Electronics Cleaner	6%	0%	0%
Fire Extinguisher	1%	0%	0%
Other Applications	12%	3%	14%
Test Properties	36%	41%	29%
Thermodynamic Properties	32%	38%	43%
Vapor Compression Cycles	38%	45%	57%
Materials Compatibility	32%	45%	0%
Reactivity	12%	14%	0%
Environmental Impact	5%	0%	0%
Other Properties	15%	17%	14

3.3.1 HFC-134a

The compound HFC-134a was frequently cited as a replacement for CFC-12, which is currently used extensively in refrigeration, water chilling, and car air-conditioning applications. Therefore, it was not surprising that of those investigating HFC-134a, almost 90% are interested in refrigeration and air-conditioning applications. Just under half of these respondents are involved in optimization studies of the vapor-compression cycle when using HFC-134a. About half of the researchers are also involved in examining materials compatibility issues, and half are involved in testing to establish thermodynamic and chemical properties of HFC-134a. Table 3.4 lists the purposes of the research into this specific compound.

The specific materials compatibility research issue cited most frequently by researchers was finding appropriate lubricants for HFC-134a vapor-compression cycle equipment. One respondent is also conducting compatibility tests with metals. Another respondent is reportedly developing a test apparatus to measure thermodynamic properties (presumably, also applicable to testing of other alternatives). However, because most researchers reported looking at applications and materials compatibility issues, the majority seem to have either concluded thermodynamic property testing or have access to thermodynamic data collected by others.

Other HFC-134a research areas that were mentioned by at least two respondents also relate to vapor-compression refrigeration cycle applications, and include 1) testing the suitability of desiccants, 2) optimizing refrigeration compressor designs, and 3) conducting heat transfer tests to establish heat exchanger performance and transport properties. Finally, one respondent is looking at the feasibility of retrofitting CFC-12 systems for use with HFC-134a, and another is conducting long-term aging tests of vapor-compression cycle equipment that uses HFC-134a.

3.3.2 HCFC-22

Of those respondents who indicated HCFC-22 as the main compound they are studying, most are studying vapor-compression equipment applications. Perhaps because this compound is already widely used in residential and commercial applications the respondents seemed to be in relatively advanced stages of research. One respondent indicated that a CFC-12 system had been converted to HCFC-22 "at a reasonable cost," and one had conducted experimental tests of a HCFC-22/HCFC-142b binary mixtures in refrigerators, yielding results that are "very satisfactory." Other respondents are conducting thermodynamic calculations of ternary blends. Table 3.4 lists the purposes of the research into this specific compound.

3.3.3 Other Compounds

A number of specific alternatives were mentioned by either one or very few respondents. Listed below are those alternatives, the country conducting the research, the purpose of the research, and any problems with the alternative.

Aqueous Cleaning. A respondent from Japan and one from Switzerland are looking at using an aqueous cleaning solution to replace CFC-113 as an electronics cleaner. Specifically, the Japanese respondent has

> replaced CFC-dependent draining and drying technology with a dipping and drying device that used distilled water, and...[he is] investigating the use of spin drying and other existing technologies.... [The respondent is] studying the use of washing technology that uses water-based solvents instead of CFCs...[and is] developing substitutes for CFCs used for dilution purposes in surface treatment processes.

The Swiss respondent is looking at the environmental impact of aqueous methods for electronics cleaning. One challenge he noted with this application was the cost for waste water treatment because water cleaning requires "different equipment, dryers, and waste water treatment."

Azeotropic Mixture. A respondent from India is investigating an azeotropic mixture to replace CFC-12 in refrigeration equipment. The respondent is analyzing thermodynamic properties, vapor compression cycles, and material compatibility issues. In the respondent's opinion, the cost and lack of the potential azeotropic compounds he is studying are minor problems.

Ethylchloride. A respondent from Austria is investigating ethylchloride as a replacement for CFC-22 in heat pumps for refrigeration applications. The respondent is studying the thermodynamic properties, vapor compression cycle analyses, materials compatibility, and reactivity issues. Problems include technology modifications to address compatibility problems with aluminum, environmental issues, and limited applications.

H2O-LiBr. A respondent from France is analyzing a water/lithium/bromide mixture as a replacement for CFC-12. He is currently in the "first year of study of a 4-year development plan." Specifically, he is investigating the potential of using a water/lithium bromide mixture as a refrigerant by studying the materials compatibility issues and its potential for what he referred to as a "hybrid cycle" that combines both the absorption and vapor-compression refrigerant cycles. A simulation program and bench study have been completed. Some problems he cited include the many technology modifications required to develop the hybrid cycle, its high expected first cost, and its limited applicability.

Binary Mixtures. Three respondents from Canada are working together to analyze various two-component mixtures of HFCs and HCFCs for applications in refrigeration systems. They are analyzing both the thermodynamic properties and the vapor-compression cycle efficiency of the binary mixtures. They hope to develop compounds that require little or no modifications to existing air-conditioning equipment (i.e., a "drop-in" substitute).

HCFC-123. A respondent from Japan is looking at HCFC-123 as a replacement for CFC-11 as a foaming agent for insulation in refrigerators. His goal is to make refrigeration insulation with the same insulating value and cost as CFC-11, but he is finding it "very difficult." The insulation he has tested, which is made using HCFC-123, lacks "high quality."

A respondent from Italy reported testing the use of HCFC-123 in geothermal power cycles. The respondent has "measured the vapor pressure and performed some preliminary test[s] on thermal stability up to 200°C." The respondent notes that "in [the] system (a hot-water power cycle), changing from CFC-11 to HFC-123 should not pose real problems, except...some compatibility problems with materials [may] arise."

HCFC-141b. Two respondents, one from Korea and one from Taiwan, are investigating HCFC-141b as a replacement for CFC-11. The respondent from

Korea is also studying how to manufacture HCFC-141b in large quantities (i.e., an "industrial manufacturing process of HCFC-141b"). The Taiwanese respondent is testing the application of the compound as an electronics cleaner and insulation/blowing agent. Some of the problems with using HCFC-141b that were cited by these respondents include its higher cost, flammability, required technology modifications, limited applications, and legal issues. The respondent is planning to study how to manufacture the alternative.

HFC-152a. Individual respondents from Canada, Germany, and the United Kingdom are looking at HFC-152a as an alterative to CFC-12. All three respondents are studying the applications of the compound in vapor-compression cycle refrigeration systems. The respondent from Canada is also testing its applicability as a foaming agent for insulation. All respondents mentioned that the major problems with the compound are its flammability and the associated product liability issues.

The respondent from Canada noted, "We believe that the alternate will be viable, assuming materials compatibility (as it effects long-term reliability) doesn't become an issue." Future plans include completing long-term life cycle testing and field trials.

The primary goal of the German respondent's research is to test thermodynamic properties, analyze HFC-152a vapor-compression refrigeration cycles, and identify and resolve materials compatibility issues. The German respondent noted that because the alternative still has 1% GWP (however, no ozone depleting potential), environmental issues are still a concern. He further stated that lubricity, lack of supply (the compound is a byproduct of an oil cracking process), and the limited applications of HFC-152a are issues, but of secondary importance. Research funding in Germany, in his opinion, is a moderately serious problem at this time. The respondent noted that "several projects funded by the German government will end during this year. The money from the German government will be spent mainly for projects in East Germany."

Mixture of HFC-152a and HFC-134a. One of the respondents from Germany is looking at a mixture of HFC-152a and HFC-134a to replace CFC-12. The primary goal of his research is to investigate the thermodynamic properties. He noted that lubricity is a moderately serious problem.

Hydrogen. A respondent from Germany is investigating hydrogen as a propellant that is stored in a metal-hydride compound as a replacement for CFCs in aerosol spray cans. Specific activities include tests to determine "dependence of aerosol quality on propellant pressure...and leakage of propellant out of commercially available spray cans." Technology issues needed to further this application include "higher [working] pressures, modified [spray] nozzles, and the need for additional metal-hydride storage, manufacturing, and recycling (reclaiming)."

Isobutano. A respondent from Mexico is examining the use of "Isobutano" as a replacement for CFC-11 in aerosols. He indicated that technology modifications are a moderately serious problem and flammability is a very serious problem. "Aerosol filling technology must be modified to take into account flammable substitutes."

Nonazeotropic refrigerant mixtures (NARMs). The use of NARMs (e.g., HFC-22/HFC-142b) as potential replacements for CFC-12 is being studied by a Swedish respondent. The primary goal of the research is to test the application of the NARM in heat pumps by conducting vapor compression cycle analyses and testing heat transfer properties. A significant technical challenge is to develop a suitable evaporator design for the two temperature evaporative process that are characteristic of a NARM. Future plans include "testing full-sized heat pumps [and] the evaporative heat transfer process, modeling [NARMs] with oil, and [then] refining cycle calculations."

Ammonia. Ammonia, long used in industrial refrigeration processes worldwide, is currently being studied in Norway, Sweden, and the United Kingdom for wider commercial air-conditioning and refrigeration applications.

The Norwegian respondent is studying the compound as an alternative for both CFC-12 and CFC-502. The primary goal of his research is to analyze vapor compression cycles at temperatures and pressures typical for commercial refrigeration equipment. "A prototype of a compact ammonia refrigeration unit...is under construction." Because ammonia is not compatible with copper, the most common heat transfer material used in refrigerant heat exchangers, selection of new construction materials will be necessary. Other needed technical modifications include implementation of a method to prevent ammonia gas from escaping into occupied spaces. Finally, a basic problem hindering the use of ammonia is the "lack of ammonia tradition and experience in some countries."

The respondent from Sweden is studying "ammonia in mechanical refrigeration combined with liquid nitrogen and carbon dioxide" for industrial food freezing.

The United Kingdom respondent has made an assessment of necessary plant changes and provided guidance on the safe use of ammonia. The main issue associated with current ammonia-based technologies "is providing [adequate] safety systems when [they are] used for building air-conditioning applications." The most serious obstacle in his view is "lack of industry skills in using [ammonia] and an aversion of [the air-conditioning] industry and users."

Water. A respondent from Germany is studying using water at applications above 0°C as a replacement vapor-compression cycle refrigerant for CFC-11, CFC-12, and CFC-114. Though the specific uses for which a water-based system could be applied were unclear from the survey answers, the respondent indicated that a large-scale plant that uses water as the refrigerant has been completed, and is now a commercially available technology. He did indicate he will continue to improve the components of the system.

Ternary Blend (HCFC-22, HFC-124 and HFC-152a). A respondent from The Netherlands is investigating the DuPont "ternary blend" as a replacement for CFC-12 in refrigeration equipment. He noted that flammability is a possible problem.

ISCEON 69S. A respondent from the United Kingdom has developed a non-CFC refrigerant that is now on the market called "ISCEON 69S," a blend that includes HCFC-22, propane, HFC-218, and is marketed as a replacement for CFC-502 in refrigeration equipment. The respondent indicated that no technology modifications are necessary to existing CFC-502 equipment, which would make this one of the few potential "drop-in" alternatives.

3.4 ISSUES

About half of the respondents reported that modifications to technologies currently using CFCs would be required to use the alternative compounds they were studying. That is, there is no direct "drop-in" non-CFC alternative refrigerants. The most frequently cited issue associated with the transition to CFC alternatives was "technology modifications." One third of these respondents qualitatively rated technology modifications as a moderately serious issue.

Cost of the alternatives and of their implementation was the second most frequently mentioned issue from the list, with most of the respondents indicating it to be either a moderately serious or minor problem. Finally, just under half of all respondents said lubricity is a problem, with most claiming it to be a moderately serious problem. Table 3.5 lists the problems associated with the alternative research.

TABLE 3.5. Specified Problems/Issues Identified With the Research

Problem/Issue	All Respondents	HFC-134a Respondents	HCFC-22 Respondents
Cost	61%	93%	86%
Environmental Issues	33%	48%	43%
Technology Modifications	63%	86%	72%
Limited Applications	38%	45%	57%
Lack of Supply	37%	52%	43%
Lubricity	48%	79%	43%
Efficiency	37%	62%	43%
Legal Issues	28%	35%	29%
Funding	31%	38%	43%
Other	21%	10%	14%
None	1%	0%	0%

3.4.1 HFC-134a

Three quarters of the respondents studying HFC-134a specifically indicated that technology modifications to existing CFC vapor-compression cycle equipment would be required in order to use HFC-134a as an alternative refrigerant.

As can be expected given the large number of respondents researching HFC-134a/oil mixtures, over half of the respondents replied that oil compatibility was an issue. Several other respondents mentioned that the heat transfer equipment used in vapor-compression cycle equipment would need modification to account for the heat transport properties of HFC-134a. Other issues related to using this alternative in current CFC vapor-compression cycle equipment include refrigerant desiccant effectiveness; design of components such as tubes, seals, and hoses; and use of materials such as plastics, metal and copper. Individual respondents also mentioned (but did not elaborate upon) problems with compressor-motor insulation compatibility in hermetic systems, energy efficiency, reliability, electrical resistivity, and leaking.

As shown in Table 3.5, the most frequently cited issues associated with the use of HFC-134a were its cost, technology modifications, and lubricity. Of these three, most of the respondents indicated the severity of these problems was "moderate." About half of the respondents indicated as problems the lower operational efficiency of HFC-134a relative to (presumably) CFC-12, its lack of supply, environmental issues, and its limited application. Less frequently mentioned problems include legal issues and difficulty in obtaining research funding.

3.4.2 HCFC-22

Increased cost was the most frequently mentioned problem indicated by the respondents who are researching HCFC-22. This finding seems at first surprising, for HCFC-22 is commonly used in many refrigeration and air-conditioning applications, and thus would be expected to be relatively inexpensive to adapt to similar applications where CFCs are currently used. It is, however, about twice as expensive to manufacture as, for example, CFC-12 or CFC-11. In addition, a vapor-compression HCFC-22 cycle operates at higher pressures for the same temperatures than a similar CFC-12 cycle, which would increase the production cost of system components. The cycle efficiency of HCFC-22 is also lower than CFC-12 or CFC-11, resulting in higher energy usage for the same capacity. Finally, its different refrigeration effect and operating pressures clearly would require redesign of compressors and heat exchangers in refrigeration applications that use CFCs, as well as extensive retooling of manufacturing facilities. However, because operation of HCFC-22 in air-conditioning and refrigeration is, as already stated, well understood, more innovative technology modifications to adapt it to applications as a replacement for CFCs do not appear to be necessary. As one survey respondent said "design modifications are necessary but on the same technology base."

"Limited applications for HCFC-22" was also mentioned as a problem. However, no details were given in the surveys

3.5 RESEARCH PLANS

The responses varied to the question inquiring about future research studies. Several respondents plan to study heat transfer in the condensation and evaporation of refrigerant mixtures. Other studies include studying alternative refrigeration cycles to the vapor-compression cycle (e.g., Brayton cycle), redesigning centrifugal chillers, and developing compressors that are optimized for use with alternative refrigerants. Other general research plans include the following: studying ways to improve CFC containment to minimize losses to the environment, recycling existing CFC supplies to extend their useful life, determining the suitability of alternatives as "drop-in" substitutes in existing CFC-using refrigeration systems, studying flammability issues relating to alternatives, improving energy efficiency in air-conditioning equipment, studying foam blowing, and researching the condensation and evaporation of NARMs. One respondent is also beginning tests using a new type of CFC recovery/recycling machine. The overall objective of current and future research being conducted by one respondent from the United Kingdom is "to assess the implications of the CFC phaseout for the U.K. building industry, with particular emphasis [on] providing guidance on new [alternative] refrigerants, changes to performance, and costs, including equipment conversion."

It is interesting to note that of those respondents who indicated that HFC-134a offers the most potential for alleviating the CFC problem, almost all of the respondents said that their future plans entail studying other alternatives. This finding suggests that HFC-134a may not be an appropriate alternative to CFCs in some applications. Of these respondents, most indicated that they will be investigating alternatives to HCFC-22. A few expect to be evaluating alternatives to R-502 (an approximately 50/50 blend of HCFC-22 and CFC-115) and other alternatives to CFC-11. Some promising alternative fluids these researchers expect to be studying include ammonia (NH_3) and HFC-152a.

3.6 RESEARCH THAT SHOULD BE PURSUED

Table 3.6 lists other groups and organizations, specified by the respondents, that are conducting research into CFC alternatives. When asked what type of research should be pursued by these groups, several respondents indicated additional theoretical vapor-compression cycle analyses should be performed. Several respondents also felt that the flammability issues with some of the alternatives needed to be addressed. A few mentioned the need for further study of the potential environmental effects of CFCs including global warming and ozone depletion. Retrofitting the existing CFC systems for alternatives, developing drop-in alternatives, and conducting long-term reliability tests of systems using the alternatives were also mentioned. One respondent mentioned the need for looking at low-cost production methods. Finally, a few respondents also mentioned the need for increased international cooperation and "intentional" collaboration in the study of CFC alternatives.[a] This final suggestion could be quite advantageous given that the research progress between various countries appears to differ significantly.

(a) One example of an international on-going collaborative research activity is the "Alternative Fluorocarbons Environmental Acceptability Study" (AFEAS) (1991). This work is co-funded by 12 of the world's chemical companies.

TABLE 3.6. Other Groups/Organizations Conducting CFC Research

Country	Groups/Organizations
Austria	Forschungrzentrum Seikersdorf Aubeninstitut, Techn Univ., Vienna
Brazil	All big refrigerator companies under ABINEE (Industry association)
Germany	DKV (German Refrigeration and AC Society - Deutscher Kalte- UND Klimatechnischer Verein, Pfaffenwaldring 10, D-7000 Stuttgart fax 0711/685.3242)
	Various companies (Hoechst, Basf, Kali Chemie, Huls Ag etc.)
	Universities of Hannover, Narls Ruhe, Nunchen, Dresden
Japan	Keio University Kobe University National Chemical Laboratory for Industry Freon Companies (Asahi Glass, Daikin, Showa Denko) Japanese Association of Refrigeration
Mexico	IMACC (Mexican Aerosol Institute)
The Netherlands	Heat pump Center - Sittard, NL TNO- Apledoom - NL University of Delft - Delft, NL (absorption processes)
United Kingdom	ICI Phone Pooling Dupont ICI C&P, The Heath, Runan, England National Engineering Laboratory, East Kilbride

Most of the HFC-134a respondents wanted to see more research into the thermodynamic and transport properties of other alternatives and more testing of HFC-134a/lubricant mixtures for vapor compression cycle equipment. Another material compatibility issue cited by one researcher regarded reactivity of alternatives.

3.7 FUNDING

Over half of the respondents did not answer the research budget questions, perhaps indicating that funding is a sensitive issue.

3.7.1 Funding Levels

Those that answered the funding question reported a wide range of funding levels, ranging from $1,000 to $4 million U.S. dollars. Because the question asked for the research budget for 1990, the exchange rates for the first working day of 1990 were used for this analysis. Table 3.7 lists the exchange rates for each country. West Germany's rate was used for Germany's exchange rate. Table 3.8 shows the total dollar amount (in U.S. dollars), the median amount, the range of reported budgets, and the number of respondents for each country. Japan, Germany, Canada, and the United Kingdom had the greatest number of respondents answering the question. However, the range and the median amounts widely varied. Norway and Austria each had one respondent who answered the question, yet they had relatively large budgets.

3.7.2 Funding Sources

As shown in Table 3.9, about half of the respondents receive their funding from industry, followed by government, other sources, and universities. A few said funding would decrease, while most said funding would remain stable over the next year. With a few exceptions, similar trends were found when the data were analyzed by specific compound and by country. The German responses to the stability of the funding level were mixed: 40% thought that funding would decrease in the next year; however, 30% felt that it would remain stable. Over one-third of the United Kingdom respondents felt funding would decrease. Of the five Canadian respondents that answered the funding source question, four indicated they receive funding from the government, while two indicated industry.

3.8 RESEARCH DISSEMINATION

Presentations at technical conferences were the most common method cited by respondents for learning the research results of others. Journals were the second most common method. In Canada, professional organizations are the most common method for learning of research by others.

TABLE 3.7. Exchange Rates, January 2, 1990
(Wall Street Journal 1990)

Country	Exchange Rate
Australia	0.784
Austria	0.8294
Brazil	0.11495
Canada	0.8613
China	0.211774
Denmark	0.15
France	0.1707
Germany (West)	0.5834
India	0.05952
Italy	0.0007777
Japan	0.006825
Korea	0.0014684
Mexico	0.0003735
New Zealand	0.593
Norway	0.1506
Sweden	0.1605
Switzerland	0.6315
Taiwan	0.038505
The Netherlands	0.5166
United Kingdom	1.605

TABLE 3.8. Research Budgets by Country

Country	Total Budget In U.S. $	Median	Range	Respondents
Australia	414,700	414,700	414,700	1
Austria	78,400	78,400	78,400	1
Brazil	300,000	300,000	300,000	1
Canada	568,458	142,115	68,000-215,325	4
China	1,000	1,000	1,000	1
Denmark	320,000	160,000	20,000-300,000	2
France	341,400	170,000	85,350-341,400	2
Germany	1,502,255	116,680	14,585-1,166,800	5
India	5,952	5,952	5,952	1
Italy	40,000	40,000	40,000	1
Japan	6,590,225	568,250	20,475-4,000,000	6
Korea (South)	300,000	300,000	300,000	1
Mexico	0	0	0	0
New Zealand	0	0	0	0
Norway	753,000	753,000	753,000	1
Sweden	530,000	265,000	200,000-330,000	2
Switzerland	63,150	63,150	63,150	1
Taiwan	0	0	0	0
The Netherlands	250,000	250,000	250,000	1
United Kingdom	170,130	36,915	16,050-80,250	4

TABLE 3.9. Funding Sources

Funding Source	Respondents
Government	30%
University	6%
Industry	48%
Other	15%

4.0 CONCLUSIONS AND RECOMMENDATIONS

The results of the survey provide a current overview of international research activities into alternatives for CFCs.

4.1 CONCLUSIONS

The following conclusions can be drawn from the data:

- Length of Research. Most of the international research has been ongoing for more than 3 years, and the respondents expect to continue their research for at least another 3 years.

- Identification of Alternatives. The most prevalent alternatives being studied by the respondents were HFC-134a and, to a lesser extent, HCFC-22. Over one-third of the respondents reported the alternative HFC-134a as having the most potential for alleviating the CFC problem.

- Purpose of the Research. Most of the respondents indicated that they are currently investigating the application of one or more CFC alternatives to vapor-compression cycle refrigeration/air-conditioning equipment.

  The compound HFC-134a was frequently cited as a replacement for CFC-12, which is currently used extensively in refrigeration, water chilling, and car air-conditioning applications.

  Of those respondents who indicated HCFC-22 as the main compound they are studying, most are studying vapor-compression equipment applications.

- Issues. Three quarters of the respondents studying HFC-134a specifically indicated that technology modifications to existing CFC vapor-compression cycle equipment would be required in order to use HFC-134A as an alternative refrigerant. In addition, over half of the respondents replied that oil compatibility was an issue. Increased cost is the most frequently mentioned problem indicated by the respondents who are researching HCFC-22.

- Research Plans. Future research plans were quite varied. Several respondents plan to study heat transfer in the condensation and evaporation of refrigerant mixtures. Of those respondents who indicated that HFC-134a offers the most potential for alleviating the CFC problem, almost all said that their future plans entail studying other alternatives. The finding suggests that HFC-134a may not be an appropriate alternative to CFCs in some applications. Of these respondents, most indicated that they will be investigating alternatives to HCFC-22.

- Research That Should Be Pursued. Several respondents indicated that additional theoretical vapor-compression cycle analyses should be performed. Several respondents also indicated that the flammability issues with some of the alternatives needed to be addressed.

- Funding. The respondents reported a wide range of funding levels, ranging from $1,000 to $4 million U.S. dollars. About half of the respondents receive their funding from industry, followed by government, other sources, and universities. A few said funding would decrease, while most said funding would remain stable over the next year.

- Research Dissemination. Presentations at technical conferences were the most common method cited by respondents for learning about the research results of others.

4.2 RECOMMENDATIONS

The Montreal Protocol represents an unprecedented example of the ability of nations to cooperate and collaborate on issues of international concern. Much could be done to foster this expressed desire to cooperate on the CFC issue. Moreover, considerable interest in the results of this study was expressed by the respondents as evidenced by affirmatively answering the question whether they would like a copy of this report and by the relatively high response rate for an international survey written in English. The research community would probably welcome an expanded cooperative effort such as an international network or organization devoted solely to CFC alternatives developments. This network could be as simple as a newsletter describing current research efforts and results or as elaborate as an organization. Both types of networks would improve communication among the international CFC research community.

An issue needing further study is the development of technologies that are compatible with the new alternatives. Although this survey touched upon this issue, a more in-depth study would illuminate the entire range of technology developments, their purpose and problems, and chance of wide-spread use. Such a study could also help to identify opportunities for technology development. International collaboration in this effort could open many opportunities for applications of current or planned technology development or modification.

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APPENDIX A

QUESTIONNAIRE

1. *Do you or your organization conduct (or have you conducted) research into alternatives to chlorofluorocarbons (CFCs)?*

- ☐ Yes (Please go to Question 4.)
- ☐ No

2. *If no, can you provide names and addresses of researchers involved in this area of study in your country?*

Name: ____________________

Title: ____________________

Institution/Agency: ____________________

Address: ____________________

____

3. *In other countries?*

Name: ____________________

Title: ____________________

Institution/Agency: ____________________

Address: ____________________

____

4. *When did you begin conducting this research?*

- ☐ More than 5 years ago
- ☐ 3 to 5 years ago
- ☐ 1 to 2 years ago
- ☐ this year (1991)

5. *How long will you be continuing with this research?*

- ☐ More than 5 years
- ☐ 3 to 5 years
- ☐ 1 to 2 years
- ☐ complete this year (1991)

6. *What alternative chemical compounds have you studied and what CFCs are they supposed to replace?*

CFC/Halon	Alternative
____________	____________
____________	____________
____________	____________
____________	____________

For questions **7 through 10**, please select the <u>one</u> CFC alternative you have been studying that you feel has the most potential for alleviating the CFC problem. The alternative you will be referring to is:

____

7. *What are the primary goals of this research? Please check all those that apply.*

- ☐ To test applications of the compound
  - ☐ refrigeration/air conditioning
  - ☐ insulation/blowing agent
  - ☐ electronics cleaner
  - ☐ fire extinguisher
  - ☐ other ______________________
- ☐ To test properties of the compound itself
  - ☐ thermodynamic properties
  - ☐ vapor compression cycle analyses
  - ☐ materials compatibility
  - ☐ reactivity
  - ☐ environmental impact
  - ☐ other (please specify) ______________

8. *What is the status of your research to date? (Please include any results to date.)*

____

____

____

____

9. *If your research is application specific, will it be necessary to modify existing CFC technologies in order to use the alternative compound?*

☐ Yes
☐ No

Please explain:

____

____

____

____

10. *What are the disadvantages or problems, if any, of using this alternative compound instead of using CFCs? Please indicate the extent of the problem:*

1 = Very serious problem

2 = Moderately serious problem

3 = Minor problem

☐ cost
☐ environmental (please specify) ______________
☐ technology modifications
☐ limited application
☐ lack of supply
☐ lubricity
☐ efficiency (please specify) ______________
☐ legal
☐ funding
☐ other (please specify) ______________
☐ none

11. *What was your research budget for CFC alternatives for 1990? (Please indicate the currency)*

____

11a) From what sources does your funding come? Please specify the name of the organization.

- ☐ Government ____________________
- ☐ University ____________________
- ☐ Industry ____________________
- ☐ Other ____________________

11b) In the next year, do you expect this funding to (please check one)

- ☐ increase
- ☐ decrease or
- ☐ remain stable?

Please explain:

____

____

____

____

12. *What are your plans for future studies for alternatives to CFCs?*

____

____

____

____

13. *What other groups or organizations are involved with CFC research in your country?*

____

____

____

____

13a) What other groups or organizations are involved with CFC research in the international community?

____

____

____

____

14. *In your opinion, what type of research should these groups pursue?*

____

____

____

____

15. *How do you hear about the research results in CFC alternatives of other researchers? Please rank from the most common (1) to the least common (4) method.*

- ☐ presentations/conferences
- ☐ journals (please name) ____________________
- ☐ organizations (please name) ____________________
- ☐ other ____________________________

16. *We expect our report will be completed by September 1991. Would you like to receive a copy of this report?*

- ☐ Yes
- ☐ No

Thank you for completing this survey. Please return in the enclosed envelope by **April 30, 1991**.

APPENDIX B

COVER LETTER

Battelle

Pacific Northwest Laboratories
Battelle Boulevard
P.O. Box 999
Richland, Washington 99352
Telephone (509)

March 20, 1991

Dear :

The Pacific Northwest Laboratory is conducting a study to document foreign research activities on chemical compounds that present alternatives to chlorofluorcarbons (CFCs). The study is being done for the United States Department of Energy as part of their International Research Monitoring program. The Department of Energy plans to use this information to gain a global perspective on CFC research. Our initial research efforts identified you as a possible contact for our study.

The enclosed questionnaire was designed to gather general information about foreign research on CFC alternatives. We are interested in the types of research being conducted; the results of the research; problems encountered; and future studies that are planned. If you would like a copy of our final report please indicate as such on the space provided on the questionnaire and we will be happy to send you a copy.

We would greatly appreciate your input to this questionnaire. We would also like to receive copies of any reports you have on your CFC research, and we would like the names of other researchers we should contact. Please contact Alison Thurman at 509-376-4118 (phone) or 509-376-5484 (fax) with any questions or comments you may have about the questionnaire. Please return the questionnaire in the enclosed envelope by April 30, 1991. Thank you for your time and effort.

Sincerely,

Sean C. McDonald
Senior Research Economist

SCM:agt:mrp
Enclosure

APPENDIX C

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