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Update on Refrigerants: Past, Present and Future

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Contents

- Introduction
- History of Refrigerants (before 1930's)
- Introduction of CFCs in the 1930's
- Chemical Compositions and Nomenclature of CFCs, HCFCs, and HFCs Refrigerants
- Ozone Depletion and Change from CFCs/HCFCs to HFCs
- Global Warming and Change from HFCs to Natural Refrigerants
- Introduction to HFOs
- Summary

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Update on Refrigerants: Past, Present and Future

by Eckhard A. Groll

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- LEED Green Associate ☐
- LEED AP BD+C ☐
- LEED AP Homes ☐
- LEED AP ID+C ☐
- LEED AP ND ☐
- LEED AP O+M ☐

Course Description

- Update on Refrigerants: Past, Present and Future
  - In recent decades, the refrigeration and air conditioning industries have been in a state of flux primarily because of the phase-out of ozone-depleting CFC and HCFC refrigerants, and secondarily because of environmental concerns related to the direct global warming impacts of some of the replacement refrigerants. Due to these concerns, there is significant worldwide interest in using substances that are naturally occurring in the biosphere as refrigerants, which are considered benign to the environment and are termed “natural working fluids”. Surprisingly, many of these substances were already used as refrigerants at the dawn of the refrigeration technology in the late 1800’s. Thus, when looking at the refrigerants of the future, it is essential to understand which substances have been used in past. This presentation provides a detailed review of the past and present refrigerants, and proposes refrigerants and their respective technologies that could be used in the future. An assessment of their characteristics related to choice of one versus another, and an identification of trends set by these choices will be made.

Learning Objectives

1. Distinguish between the refrigerants that have been in used in vapor compression systems up to now
2. Describe the basic chemical compositions of refrigerants and the numbering system used to classify refrigerants
3. Explain the concepts of ozone depletion and global warming and the contribution of CFC/HCFC/HFC/HFO refrigerants to both issues
4. Identify refrigerants that are available for use in the future and the technical issues associated with them

Copyright Materials

Today's Refrigeration

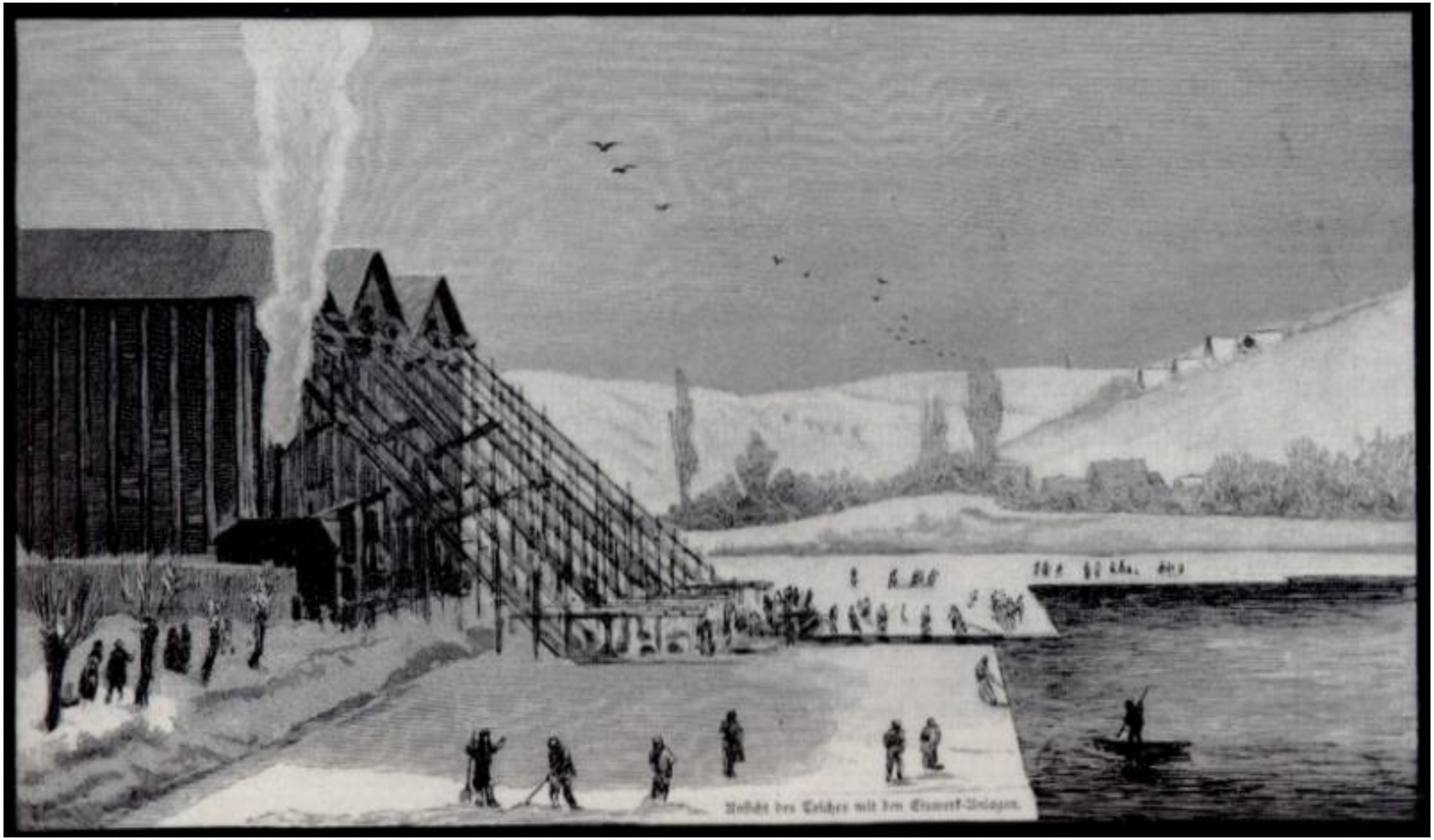
- Refrigeration of Food
- Cooling in Medical Applications
- Air Conditioning
- Cooling of Manufacturing Processes
- Chemical Engineering (Pharmaceutical and Petrochemical)
- Environmental Engineering
- Liquid Fuels for Space Applications
- Cryo Engineering
- …

→ Refrigeration is present in all aspects of engineering and the daily life

Refrigeration of the past

- How did it all start?
- At the beginning ….

“Refrigeration” from about 1600 until 1920



Harvest and transport of natural ice, e.g., from New York to Charleston starting in 1799, or up to 500,000 tons/year imported to England (1899)

Harvesting Natural Ice in 1838

Industrial Revolution in the 1800's

1834: Jacob Perkins (London) invented and patented the compression refrigeration machine, using ethyl ether. The first machine was built by John Hague in London.

1850: A.C. Twinning constructed an ether compression refrigeration system in Cleveland, Ohio (1 ton of ice per day).

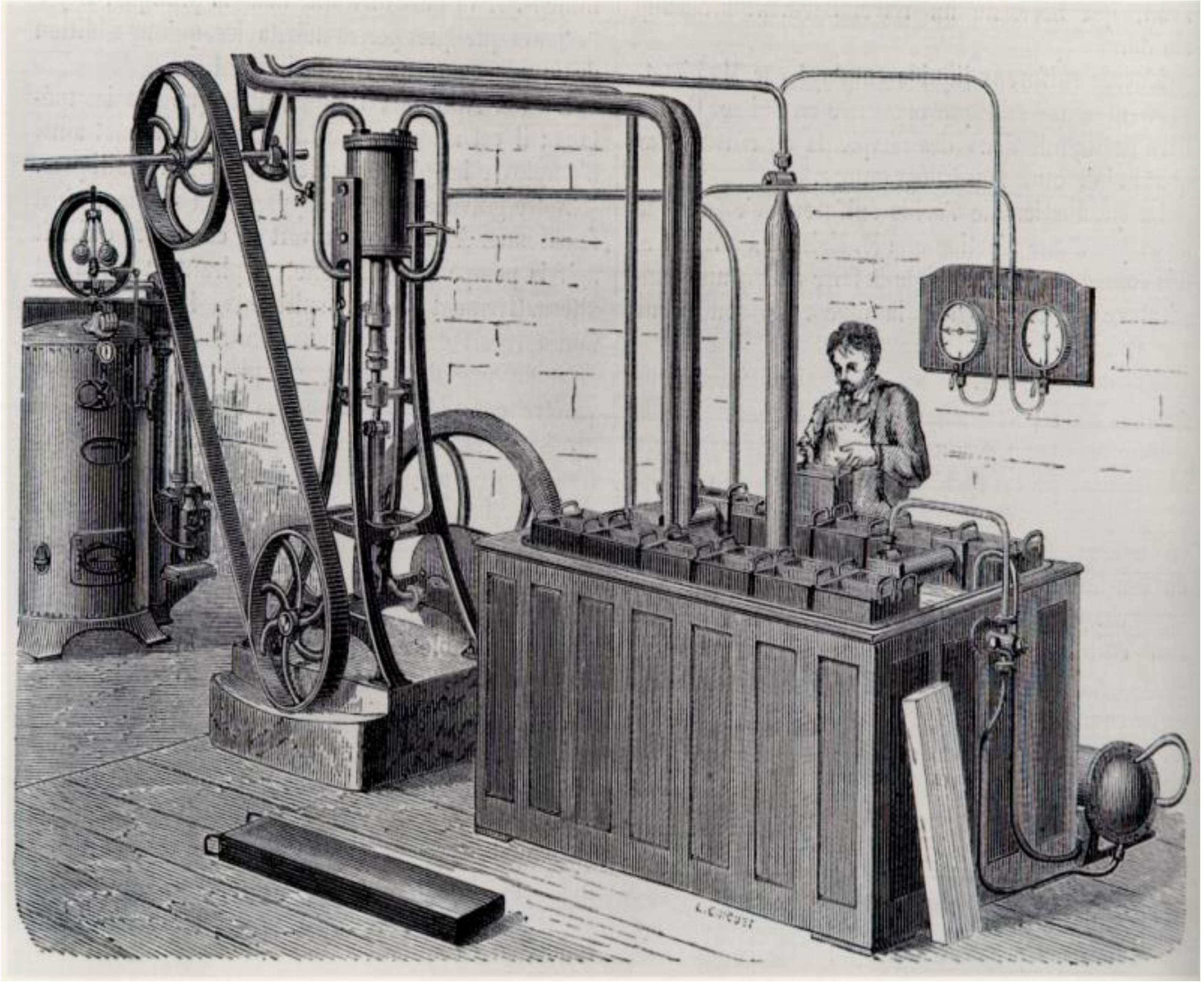
1873: David Boyle (US). First ammonia compressor.

1874: Raoul Pictet (Switzerland). First sulfur-dioxide compressor.

1876: F. Windhausen (Germany). First industrial water vapor refrigerating machine.

1887: J. & E. Hall (Great Britain). Industrial manufacture of carbon-dioxide compressors.

First Mechanical Ice Production
(Raoul Pictet 1877)



Further Developments in the early 1900's

1905: Audriffren (France). First hermetic refrigerating unit (SO_2).

1911: W. Carrier. First studies on centrifugal compressors.

1912: Maurice Leblanc (France). Prototype water vapor centrifugal compressor.

1913: W.S.E. Rolaff (US). Rolling piston compressor.

1926: General Electric (US). Hermetic compressors in domestic refrigerators.

Refrigerants up to 1930:

Substance	Refrigerant Number	Chemical Formula	NBP °C	CT °C	Rel vol	COP -15/30°C
Air [1]	R-729	-	-	-221.0	83.0	1.1
Water	R-718	H_2O	100.0	375.0	477	4.1 [3]
Carbon Dioxide	R-744	CO_2	-55.6[2]	31.0	1.0	2.56
Ammonia	R-717	NH_3	-33.3	135.0	3.44	4.76
Sulphur Dioxide	R-764	SO_2	-10.0	157.0	9.09	4.87
Diethylether	R-610	$C_2H_5.O.C_2H_5$	34.6	214.0	55.0	4.9
Dimethylether	E-170	$CH_3.\ O.\ CH_3$	-24.8	128.8	34.0	4.5
Methyl Chloride	R-40	CH_3Cl	-24.2	143.0	5.95	4.9

1. Reversed Brayton Cycle
2. Triple point
3. +5/30°C

Refrigerants up to 1930 continued:

Refrigerant	**Engineering Challenges**
Air	Inefficient; reversed Brayton cycle operation
Water	Low pressures; operation above freezing
Carbon dioxide	High pressure; transcritical operation
Ammonia	Toxic, somewhat flammable
Sulphur Dioxide	Toxic, aggressive
Ethers	Toxic, aggressive
Hydrocarbons, e.g., propane, butane, etc.	Highly flammable

Chlorofluorocarbons (CFCs)

Introduced as “Miracle Substances”

- In 1930, T. Midgley, A.L. Henne and McNary developed halogenated hydrocarbons (“Freons”) as refrigerants at the General Motors Labs for Frigidaire (Dayton, Ohio)
- Excellent thermodynamic properties
- Inexpensive, non-flammable and non-toxic
- Used as: refrigerants, solvents, propellants, blowing agents
- Stable - *perhaps too stable*

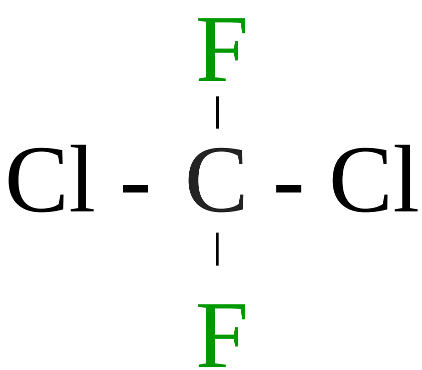
Refrigerant Designations (~~Freons~~)

R-xyz

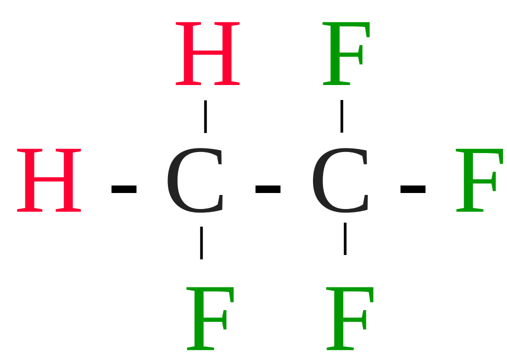
- z: # fluorine atoms
- y: # hydrogen atoms + 1
- x: # carbon atoms - 1

(remaining atoms are chlorine)

<u>R-12</u>

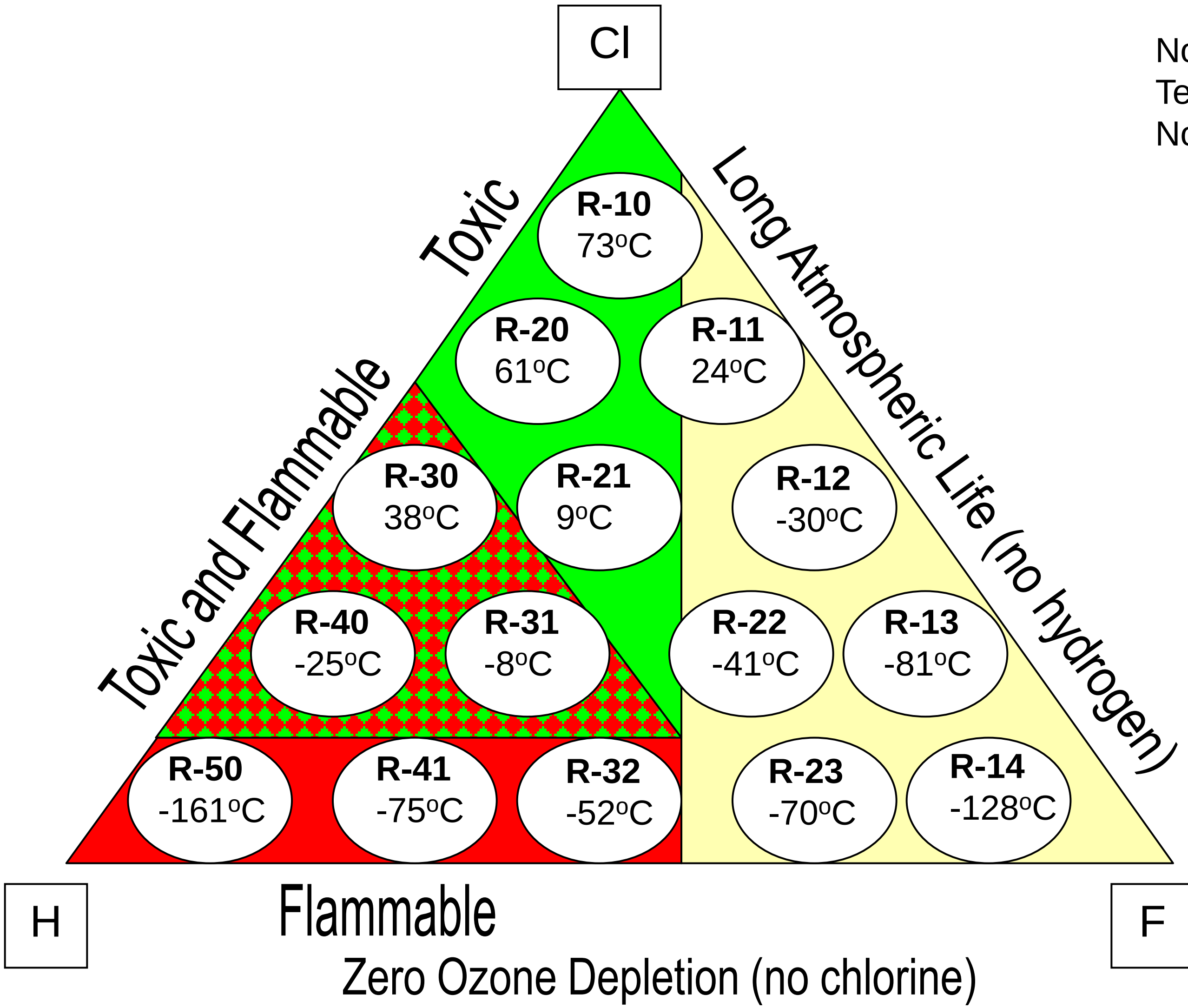


<u>R-134a</u>

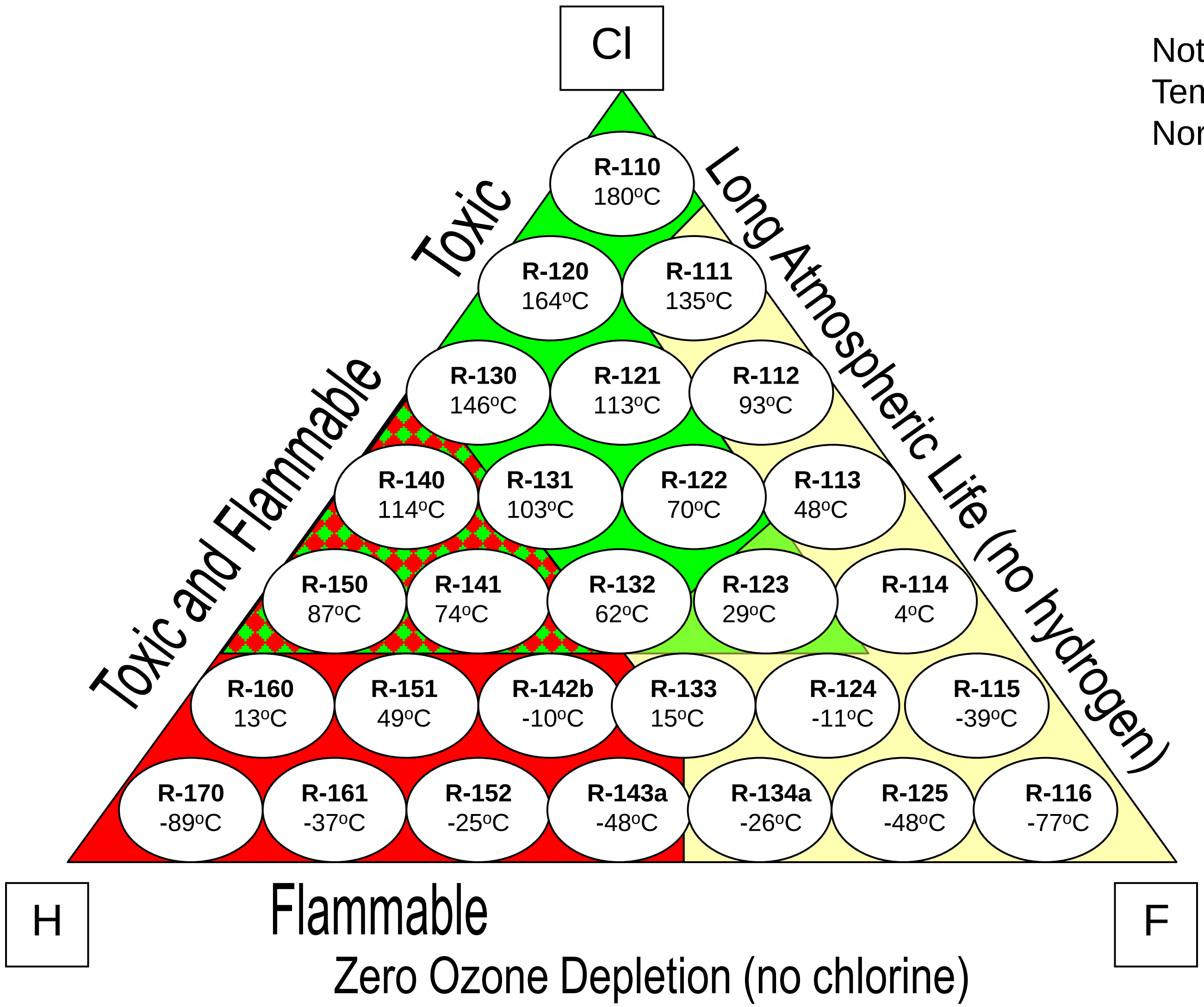


The Methane (CH_4) Family

Note:
Temperatures indicate
Normal Boiling Point

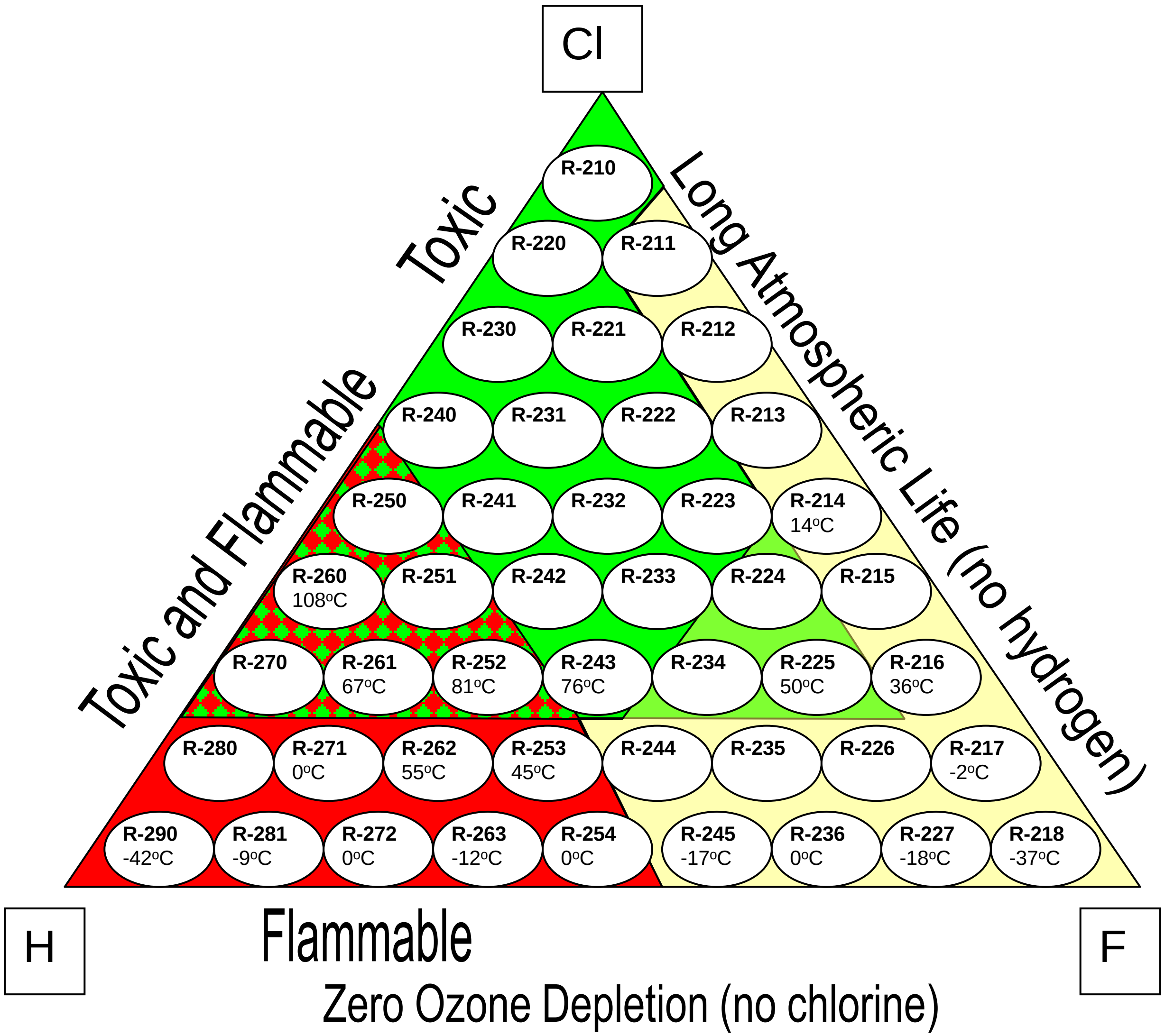


The Ethane (C_2H_6) Family



Note:
Temperatures indicate
Normal Boiling Point

The Propane (C_3H_8) Family



Note:
Temperatures indicate
Normal Boiling Point

Refrigerant Designations, continued

Zeotropic Refrigerant Blends: R-4XX

(temperature glide during evaporation & condensation)

R-404A: HFC-125/HFC-143a/HFC-134a (44/52/4 mass %)

R-407C: HFC-32/HFC-125/HFC-134a (23/25/52 mass %)

R-410A: HFC-32/HFC-125 (50/50 mass %)

Azeotropic Refrigerant Mixtures: R-5XX

(no temperature glide during evaporation & condensation)

R-500: CFC-12/HFC-152a (73.8/26.2 mass %)

R-502: HCFC-22/CFC-115 (48.8/51.2 mass %)

R-507: HFC-135/HFC-143a (50/50 mass %)

Inorganic Refrigerants: R-700+M

R-717: ammonia

R-744: carbon dioxide

Refrigerants around 1970

Household Refrigerators & Freezers | Automotive Air-Cond'g | Window Air Conditioners | Residential Air Conditioners & Heat Pumps | Commercial & Industrial Refrigeration & Air Cond'g | Large Air Conditioning

R-12 (CCl_2F_2)

R-22 ($CHClF_2$)

R-11 (CCl_3F)

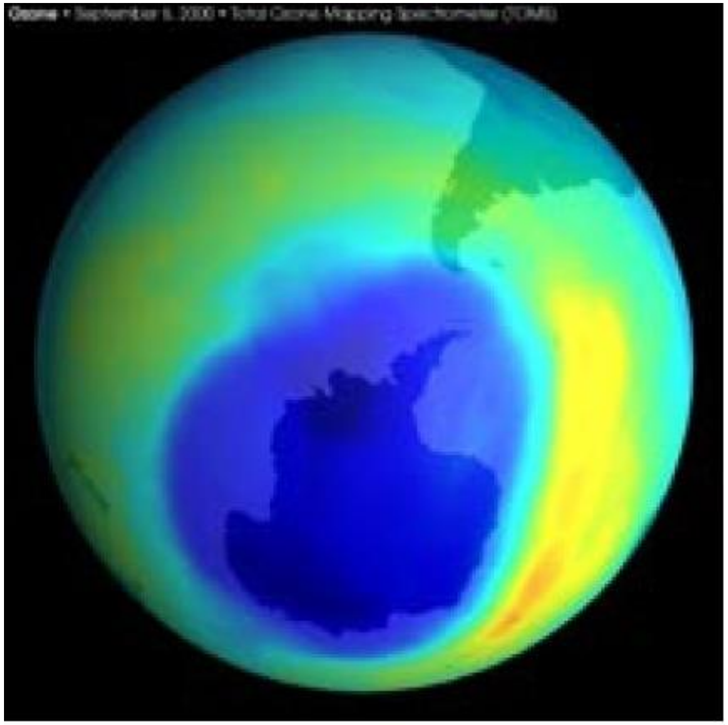
R-502 (48.8% R-22 & 51.2% R-115)

Ammonia (NH_3)

Engineering Challenges: None!

… Until …

Ozone Depletion



(Image courtesy the TOMS science team and the Scientific Visualization Studio, NASA GSFC)

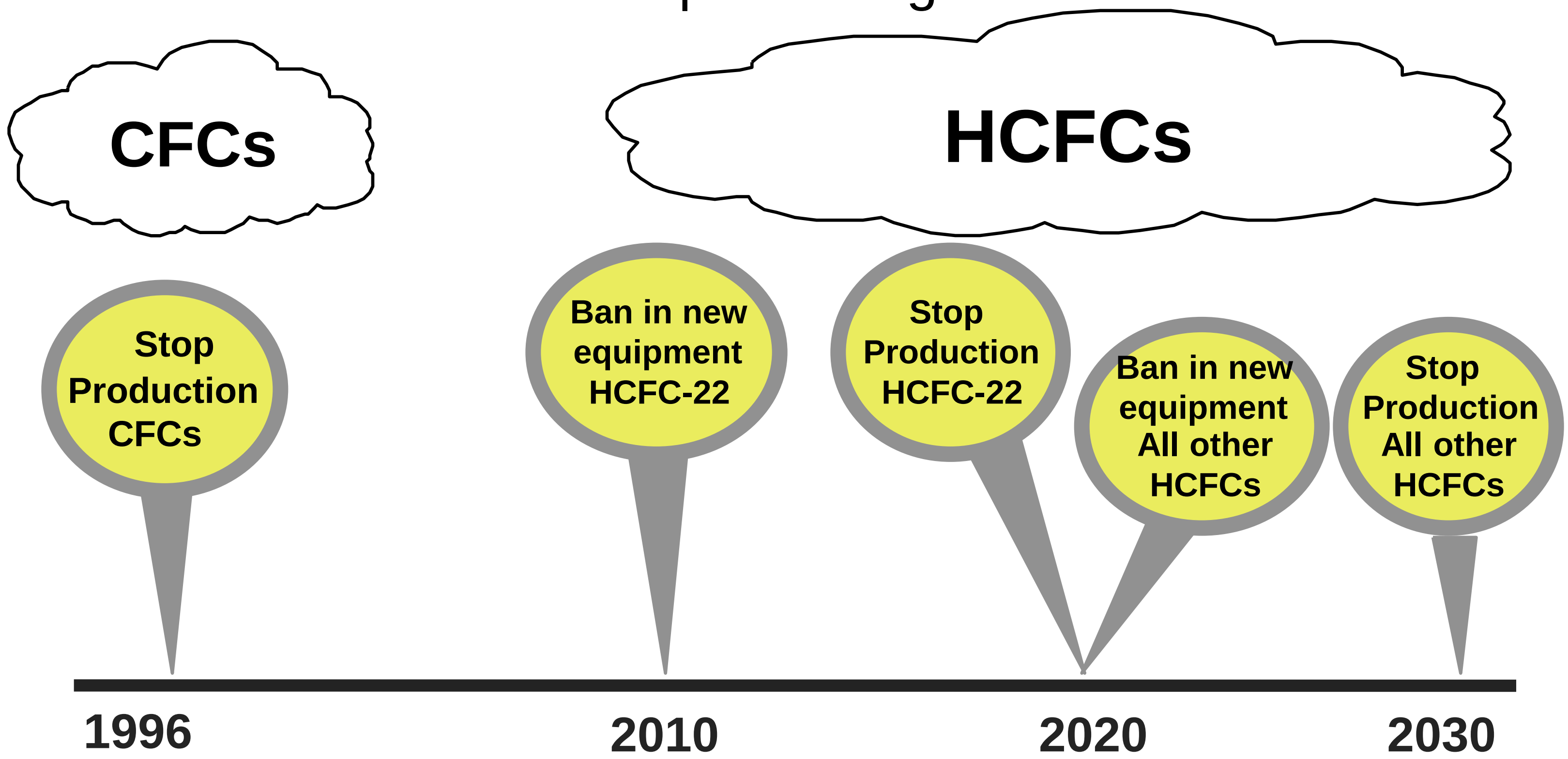
- **1974 Molina and Rowland propose ozone depletion hypothesis by CFCs**
- **1978 CFCs banned in aerosols in USA**
- **1984 First ozone hole over Antarctica**
- **1985 Vienna Convention**
  - » **Formalized international cooperation**
- **1987 Montreal Protocol**
  - » **Reduce CFC production by 50% by 1998**
- **1988 Documented losses of ozone over the Northern Hemisphere**
- **Amendments:**
  - » **1990 London**
  - » **1992 Copenhagen**
  - » **1997 Montreal**

What is Ozone Depletion?

- Normally:
  - » **7% of extraterrestrial solar radiation is UV (l < 380 nm) but less than 1% reaches earth's surface**
  - » **Dynamic equilibrium of O_3, O_2 and O radicals absorb UV radiation**
- Introduce CFC and HCFC
  - » **UV Radiation breaks off Cl from CFC and HCFC molecules**
  - » **Cl reacts with O_3 and O radicals producing more O_2**
- Net Result:
  - » **Less Ozone in the Ozone Layer**
  - » **More UV-B (280 - 320 nm) reaches earch surface**
  - » **Increase in skin cancer and cataracts**
  - » **Damage to human immunity systems**
  - » **Reduced crop yields**
  - » **Altered terrestrial and aquatic ecosystems**

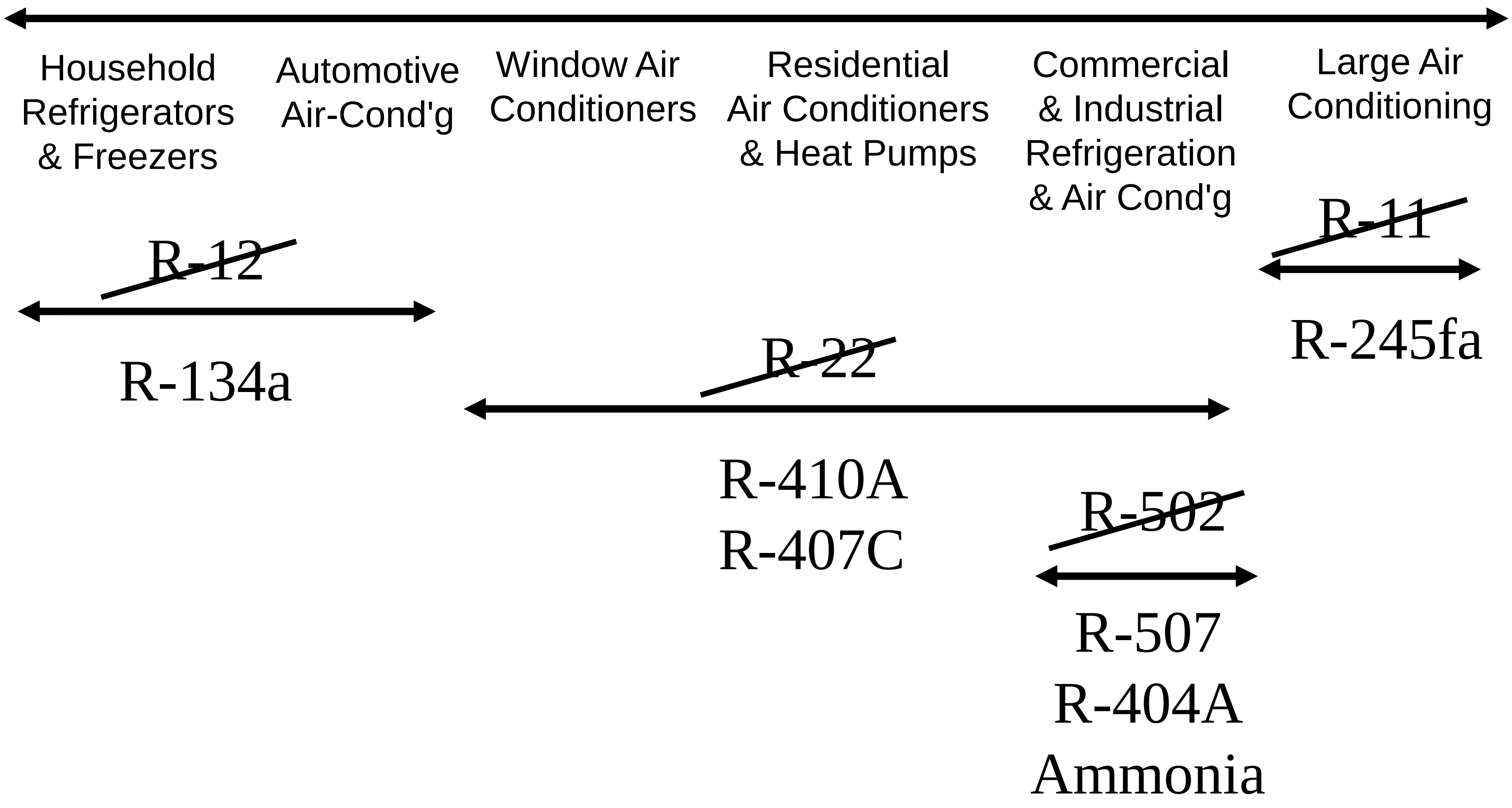
Current Phaseout Schedule

- Main Result of ozone depletion legislation:



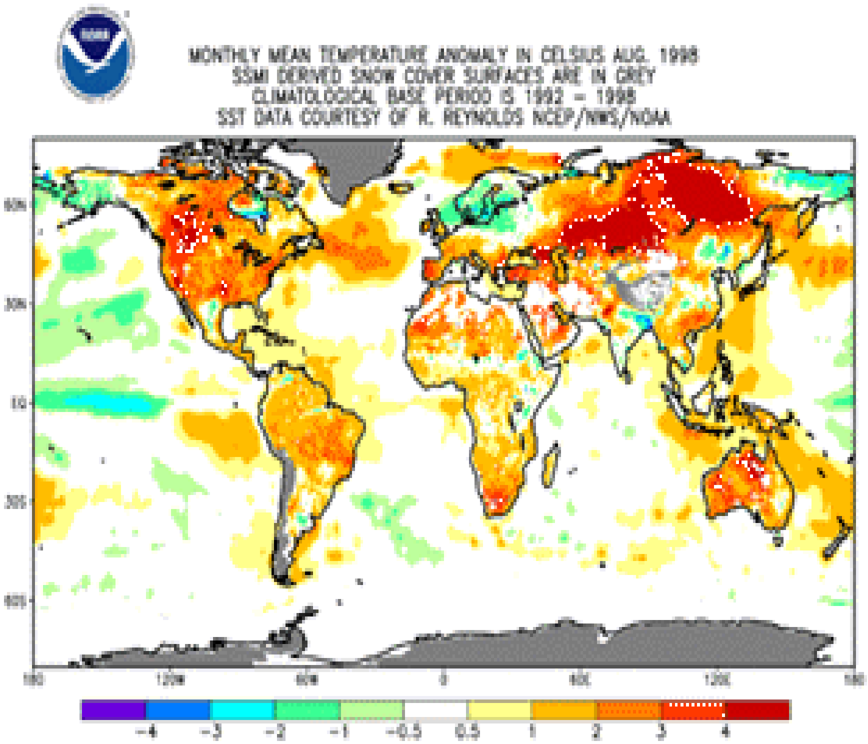
20 years later → Ozone Depletion Continues !
2005 “Ozone Hole” was one of largest and deepest ever !
Latest NASA predictions delays recovery until 2068
→nearly 20 years later than previously believed …

HFC's to the rescue!



However, ozone depletion is not the whole story!

Global Warming



1827 **Fourier describes theory of the naturally occurring global warming effect (the earth is about 30 K warmer due to the natural global warming effect, i.e., average temp is +15°C instead of -15°C)**

1896 **Arrhenius predicts the increase in CO_2 emissions due to human related processes**

1900 **Mankind welcomes the additional global warming effect**

1970 **Researchers issue warnings regarding human induced global warming**

1988 **Founding of the “Intergovernmental Panel on Climate Change“ (IPCC)**

1992 **United Nations Framework Convention on Climate Change (FCCC)**

1997 **Kyoto Protocol: drastic reduction of global warming gases emissions (CO_2, N_2O, CH_4, HFC, PFC, SF_6)**

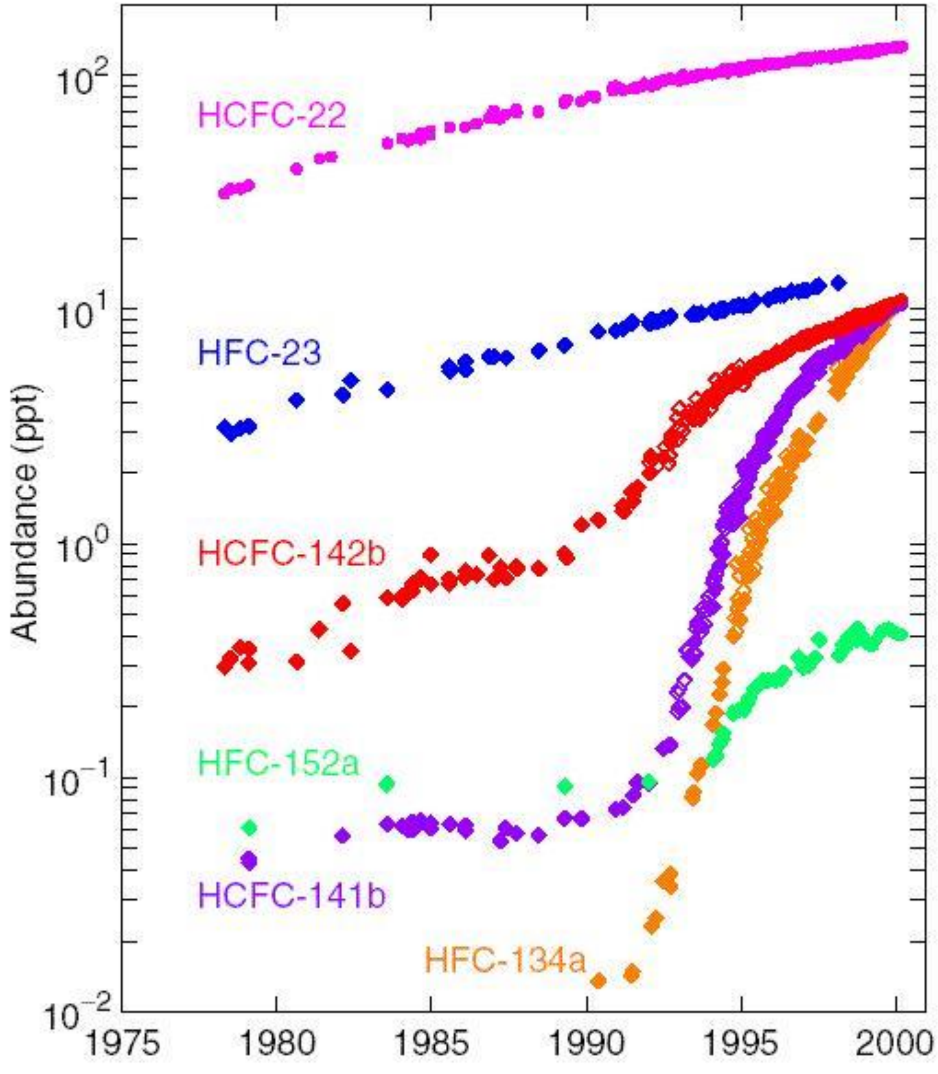
What is Global Warming?

- The earth radiates heat to the universe at various wavelengths
- Global warming gases in the upper stratosphere “close windows” of certain wavelengths
- Thus, some of the earth’s heat is not radiated to the universe, but instead reflected to the earth surface (trapped inside the atmosphere)
- The results is an average increase of the earth’s temperature
- How is the global warming impact of different substances measured?
  - » Relative to the global warming impact of the same mass of CO_2

Global Warming Impact of HFC Refrigerants

Refrigerant Number	Chemical Formula	NBP °C	Glide K	CT °C	GWP	Safety Group
R-134a	**$CH_2F.CF_3$**	**-26**	**0.0**	**101**	**1300**	**A1**
R-413A	**R-134a/218/600a**	**-35**	**6.9**	**101**	**1770**	**A1/A2**
R-404A	**R-143a/125/134a**	**-47**	**0.7**	**73**	**3260**	**A1/A1**
R-507	**R-143a/125**	**-47**	**0.0**	**71**	**3300**	**A1**
R-407C	**R-32/125/134a**	**-44**	**7.4**	**87**	**1520**	**A1/A1**
R-417A	**R-125/134a/600**	**-43**	**5.6**	**90**	**1950**	**A1/A1**
R-410A	**R-32/125**	**-51**	**0.2**	**72**	**1720**	**A1/A1**
R-508	**R-23/116**	**-86**	**0.0**	**13**	**11860**	**A1**

HFC Refrigerants in the Atmosphere



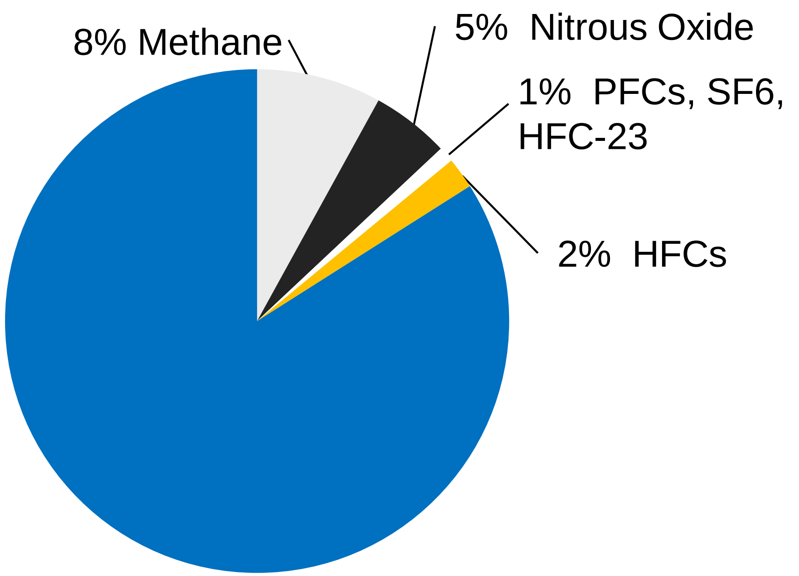
Source: NASA Measurements

Kyoto Protocol

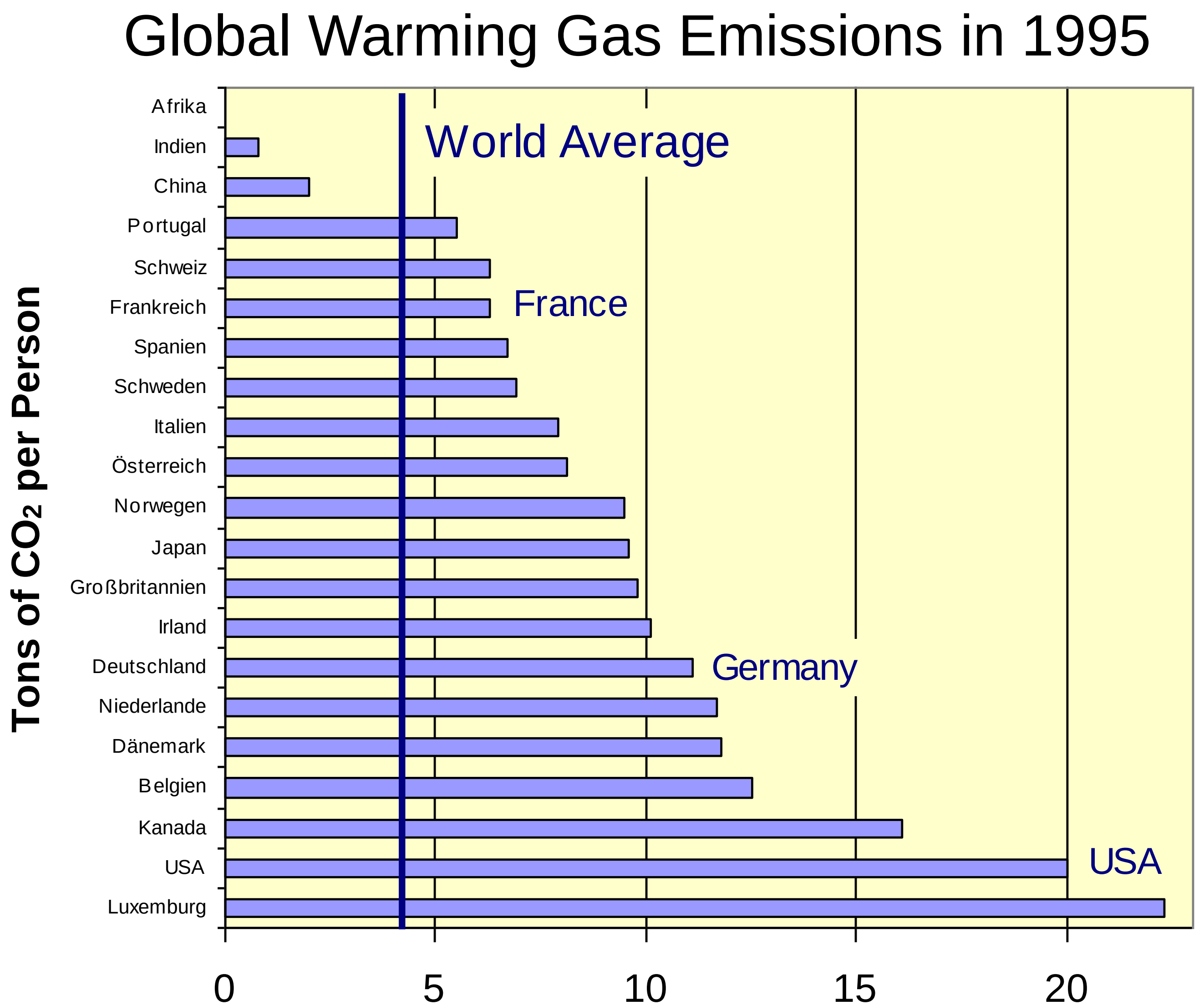
- International Climate Change Treaty
- Signed in 1997 by 187 countries
- United States of American is not currently a signatory
  - » Significant reduction commitments without a defined verification mechanism
  - » Funding for developing nations to conform
  - » Technology transfer requirements
- CO_2 from automotive and power plant emissions is the leading greenhouse gas
- HFCs are only 2% of total GHG production but seen as the most easily addressed

The Green House Gases (GHG)

Total gas emissions – assumes all gases vented to atmosphere (100% leakage)



Reduction of Global Warming Gas Emissions basend on Kyoto-Protokol



Country	Reduction of global warming gas emissions
Portugal	+ 27%
Ireland	+ 13%
Sweden	+ 4%
Norway	+ 1%
USA	- 6%
Switzerland	- 8%
EU (Total)	- 8%
Denmark	- 21%
Germany	- 25%
Luxemburg	- 28%

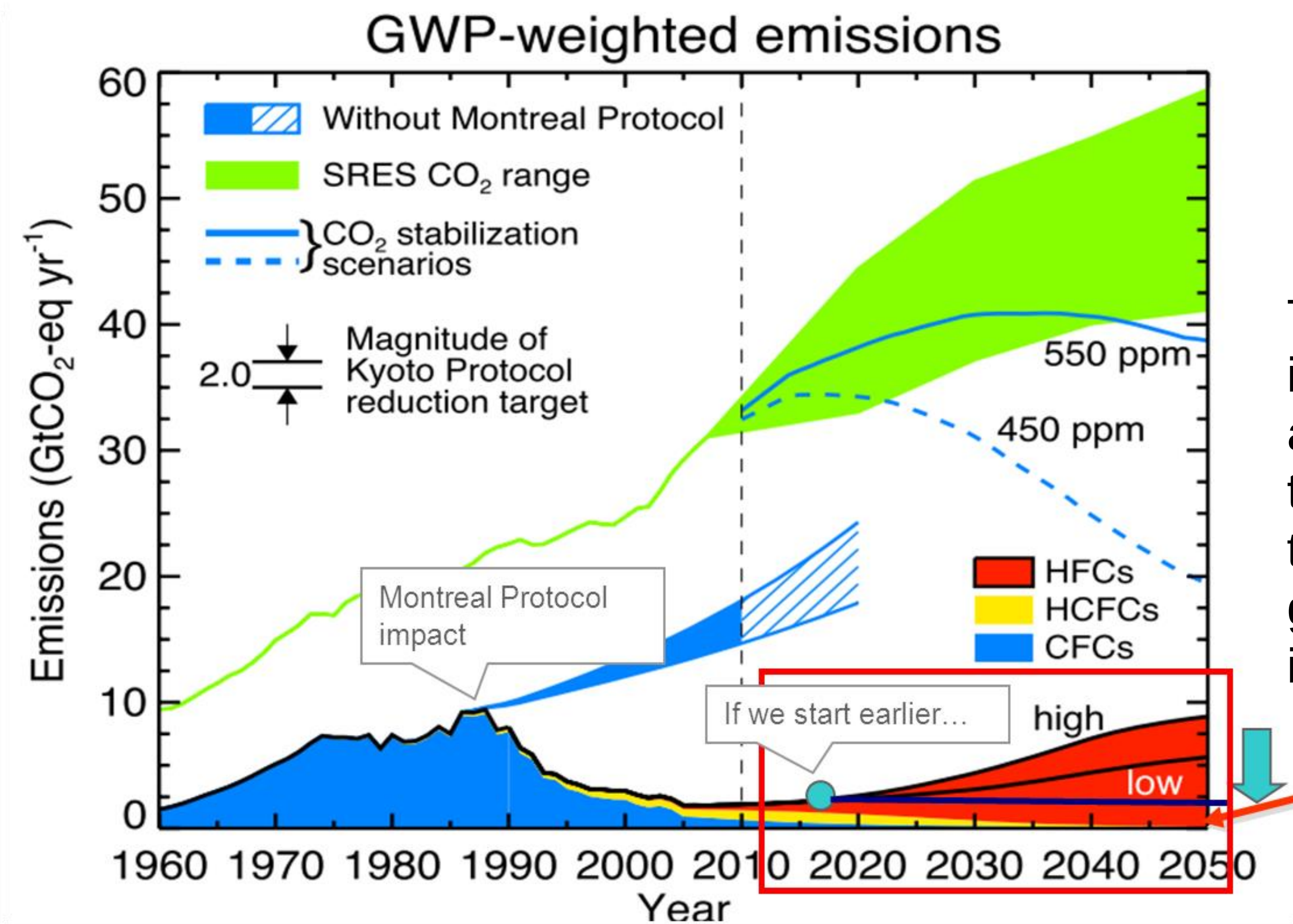
Status June 1998

CO_2 + CH_4 + NO_2 + + HFC + PFC + SF_6

HFC Impact on Global Warming

- Guus J. M. Velders et al., “The large contribution of projected HFC emissions to future climate forcing,” PNAS, July 7, 2009, Vol. 106, No. 27, pp. 10949-10954, doi: 10.1073/pnas.0902817106
  - Consumption and emissions of HFCs are projected to increase substantially in the coming decades in response to regulation of ozone depleting gases under the Montreal Protocol
  - Projected increases result primarily from sustained growth in demand for refrigeration, air-conditioning and insulating foam products in developing countries assuming no new regulation of HFC consumption or emissions.
  - New HFC scenarios are presented based on current refrigerant consumption in leading applications, patterns of replacements of HCFCs by HFCs in developed countries, and gross domestic product (GDP) growth.
  - Global HFC emissions significantly exceed previous estimates after 2025 with developing country emissions as much as 800% greater than in developed countries in 2050.
  - Global HFC emissions in 2050 are equivalent to 9–19% (CO_2-eq. basis) of projected global CO_2 emissions in business-as-usual scenarios and contribute a radiative forcing equivalent to that from 6–13 years of CO_2 emissions near 2050.
  - This percentage increases to 28–45% compared with projected CO_2 emissions in a 450-ppm CO_2 stabilization scenario.
  - In a hypothetical scenario based on a global cap followed by 4% annual reductions in consumption, HFC radiative forcing is shown to peak and begin to decline before 2050.

Global Warming: We need to Act Now



The sooner we introduce better alternatives for today's HFCs, the lower the global warming impact will be!

Source: Guus J. M. Velders et al., "The large contribution of projected HFC emissions to future climate forcing" 2009

HFC Regulations in Various Countries

Austria :	Discussion of HFC-taxes
Denmark :	HFC tax of 0.1 DKK/kg CO_2-eqivalent (e.g. R-134a = $20) HFC ban in certain applications started in 2002 and 2006
Germany :	Federal Ministry of Environment issued HFC Regulations
Great Britain :	Discussion of possible actions
Iceland :	HFC usage other than refrigeration prohibited since 1998
Japan :	Relying on voluntary leakage reduction agreements with industry
Netherlands :	Maximum leakage permitted by law
Norway :	HFC-tax 0.18 NOK/kg CO_2-equivalent effective since 1.1.2003
Sweden :	Focus on reduction of leakage Prohibition of HFC systems with more than 25 kg charge HFC-taxes 4 times higher than Denmark
Switzerland :	Measures to reduce HFC emissions
USA:	HFC recovery required by legislation

What is the Alternative?

Natural Refrigerants

Donella Meadows, American writer:

”The eighty thousand different chemicals now manufactured end up everywhere, from our stratosphere to our body. They were created to accomplish functions that can now be carried out far more efficiently with biodegradable and naturally occurring compounds.”

Ammonia, carbon dioxide, hydro carbons, water, air, helium ...

R-717, R-744, R-290, R-600a, R-718, R-729, R-704 ...

Natural Refrigerants, continued

Refrigerant Number	Chemical Formula	NBP ^oC	Glide K	CT ^oC	GWP	Safety Group
R-717	**NH_3**	**-33**	**0.0**	**133**	**0**	**B2**
R-600a	**$CH.(CH_3)_3$**	**-12**	**0.0**	**135**	**3**	**A3**
R-290	**C_3H_8**	**-42**	**0.0**	**97**	**3**	**A3**
R-1270	**C_3H_6**	**-48**	**0.0**	**92**	**3**	**A3**
R-744	**CO_2**	**-57**	**0.0**	**31**	**1**	**A1**

Consumer are Trendsetters

- McDonalds went for natural refrigerants, first HFC-free restaurant opened in Denmark in 2003
- Coca-Cola Company started to use HFC-free technology in bottle coolers in 2004
- Unilever uses HFC-free technology
- Nestlé uses natural refrigerants wherever possible
- Scandinavians value natural refrigerant supermarkets higher than conventional supermarkets
- British insurance companies are said to reduce investments in greenhouse warming industry
- Carlsberg brewing company uses HFC-free beer coolers

Available Technologies

- Iso-butane in household refrigerators/freezers in Europe
- Propane heat pumps in A, DK, D, NL, S, CH
- Ammonia and carbon dioxide in industrial refrigeration
- Ammonia water chillers for air conditioning
- Hydrocarbons in chillers
- Hydrocarbon plug-in air conditioners
- Water in chillers
- Carbon dioxide in supermarket refrigeration systems
- German car manufacturers wanted to use CO_2 in mobile AC

… and others …

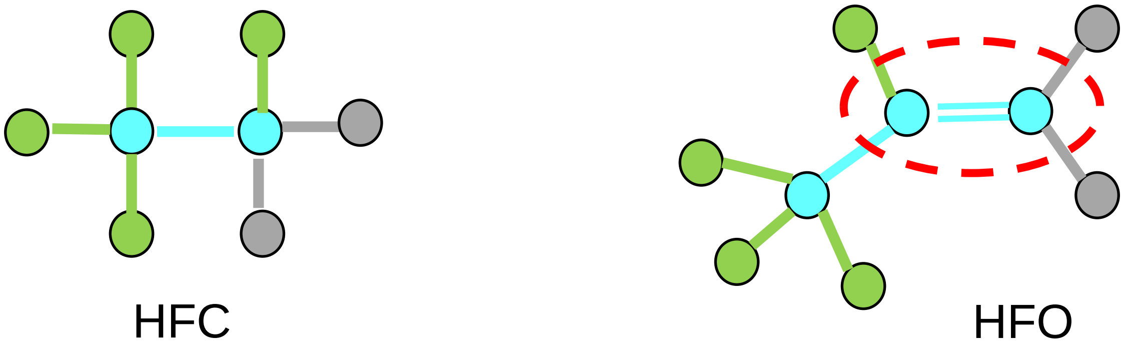
Refrigeration of Tomorrow

Household Refrigerators & Freezers	Automotive Air-Cond'g	Window Air Conditioners	Residential Air Conditioners & Heat Pumps	Commercial & Industrial Refrigeration & Air Cond'g	Large Air Conditioning
~~R-12~~ → ~~R-134a~~	~~R-12~~ → ~~R-134a~~	~~R-22~~ → ~~R-410A~~ ~~R-407C~~	~~R-22~~ → ~~R-410A~~ ~~R-407C~~	~~R-22~~ → ~~R-410A~~ ~~R-407C~~; ~~R-502~~ → ~~R-507~~ ~~R-404A~~	~~R-11~~ → ~~R-245fa~~
Iso-butane	CO_2	Propane	Propane	Ammonia, NH_3 & CO_2	Water

Have we come “full circle”?

Not so fast, what about HFO?

- Hydro-Fluoro-Olefin
- Unsaturated organic compounds also known as alkenes

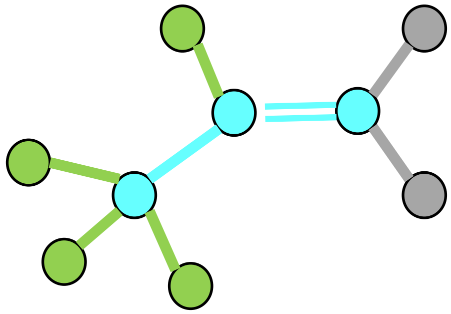


- Double bond in HFOs: quicker breakdown in atmosphere but stable in systems
- Very low GWP and zero ODP
- No or low flammability

Not so fast, what about HFO?, continued

- 2,3,3,3-Tetrafluoropropene

HFO – 1 2 3 4 y f



# of fluorine atoms

# of hydrogen atoms +1

# of carbon atoms -1

# of unsaturated bonds

Substitution on terminal methylene carbon:
a = CCl_2 ; b = CClF ;
c = CF_2 ; d = CHCl
e = CHF ; f = CH_2

Substitution on central carbon
x = Cl ; y = F ; z = H

Not so fast, what about HFO?, continued

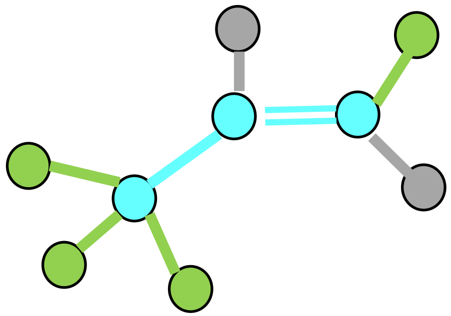
- 1,3,3,3-Tetrafluoropropene

HFO – 1 2 3 4 z e (…)

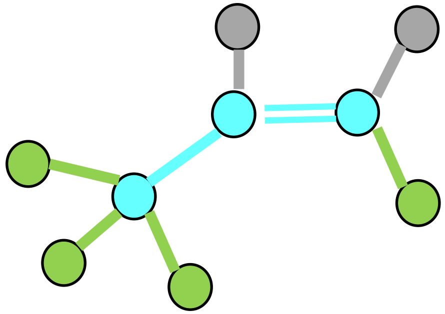
Substitution on central carbon
z = H

Substitution on terminal methylene carbon:
e = CHF

Isomer:
(E)
Trans-1,3,3,3-tetrafluoroprop-1-ene

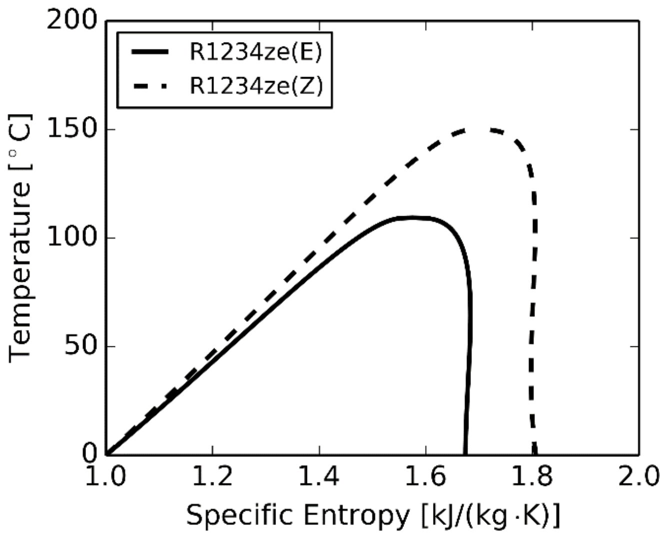


(Z)
Cis-1,3,3,3-tetrafuoroprop-1-ene



HFO Consideration

- HFO-1234yf, HFO-1234ze
  - » Classified as Mildly Flammable (A2L) by ASHRAE 34 and ISO817
  - » Use of A2L refrigerants should be discussed for wider application
- HFO-1234yf:
  - » Leading candidate to replace R-134a in mobile applications
  - » GWP of 4, no Toxicity, slightly flammable
  - » Possible applications: stationary HVAC equipment, will require significant engineering and safety code changes
- HFO-1234ze:
  - » HFO-1234ze(E)
    - – Ideal application: Foam Blowing, not necessarily HVAC&R
    - – GWP of 4
  - » HFO-1234ze(Z)
    - – High temperature heat pumps
    - – ORC applications



HFO Consideration, continued

- Trans-1-Chloro-3,3,3-Trifluoropropene or HCFO-1233zd(E)
  - » GWP of ~1
  - » ORC applications
- HFO-1336mzz(Z)
  - » ORC applications
- HFO/HFC/? Blends:
  - » Better Performance at the cost of higher GWP
  - » R448A
    - – **HFC-125 (26%) + HFC-134a (21%) + HFO-1234yf (20%) + HFC-32 (26%) + HFO-1234ze(E) (7%)**
  - » R449A
    - – **HFC-125 (25%) + HFC-134a (26%) + HFO-1234yf (25%) + HFC-32 (24%)**
  - » R450A
    - – **HFC-134a (..%) + HFO-1234ze (..%) → GWP of 547**
  - » R513
    - – **HFC-134a (44%) + HFO-1234yf (56%) → GWP of 631**

If “Mildly” Flammable becomes Acceptable?

- What about R-32?
  - » GWP: 675
  - » Suitable for all R-410A products
  - » Better performance than R-410A in cooling and heating
  - » Better performance for high ambient temp. than R-410A
  - » Classified as Mildly Flammable (A2L) by ASHRAE 34 and ISO817
  - » Refrigerant charge volume can be reduced
  - » Use of A2L refrigerants should be discussed for wider application
  - » Upper charge volume should be decided by taking into consideration safe use of multi system
  - » Continuous refrigerant containment measures are necessary

EPA Ruling from 2015

- After receiving input from industry, environmental groups, and others, EPA is approving additional low-GWP hydrocarbon refrigerants, subject to use conditions, in the following refrigeration and air conditioning applications:
  - » R170 (Ethane) in very low temperature refrigeration and in non-mechanical heat transfer
  - » R600a (Isobutane) in retail food refrigeration (stand-alone commercial refrigerators and freezers) and in vending machines
  - » R290 (Propane) in household refrigerators, freezers, or combination refrigerators/freezers, in vending machines, and in room air conditioning units
  - » R-441A in retail food refrigeration (stand-alone commercial refrigerators and freezers), in vending machines and in room air conditioning units
  - » HFC-32 (Difluoromethane) in room air conditioning units

https://www.epa.gov/snap (Significant New Alternatives Policy (SNAP) Program

Summary

- An "Unprecedented" Situation:
  - » Natural refrigerants can *not* be used in existing equipment without significant modifications
  - » Nearly all refrigeration equipment must be redesigned
  - » Alternative technologies need to improve performance, or they are not "feasible"
  - » Incentives to improve performance by better design

→ *Many Opportunities Unfold*

An "Unprecedented" Situation

- However, not this one:

Thank you!

I would be happy to answer any questions you may have!

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