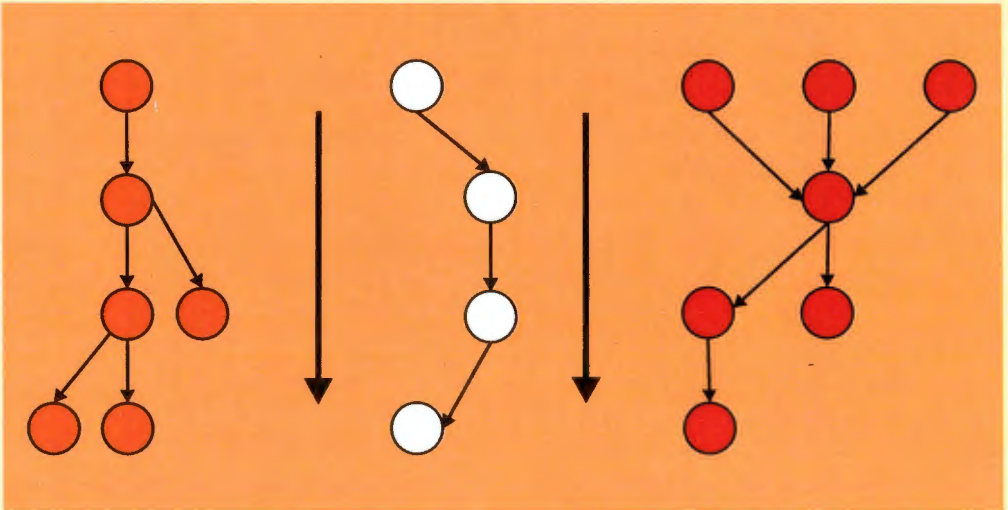


**SYSTEMS RESEARCH INSTITUTE
POLISH ACADEMY OF SCIENCES**

**MULTICRITERIA ORDERING AND RANKING:
PARTIAL ORDERS, AMBIGUITIES
AND APPLIED ISSUES**



**Jan W. Owsinski and Rainer Brüggemann
Editors**

Warsaw 2008

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This book is the outcome of the international workshop held in Warsaw in October 2008 within the premises of the Systems Research Institute. All papers were refereed and underwent appropriate modification in order to appear in the volume. The views contained in the papers are, however, not necessarily those officially held by the respective institutions involved, especially the Systems Research Institute of the Polish Academy of Sciences.

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Applications and Comparisons

Comparison of LCA Results with Those Based on METEOR. Case Study on Refrigerants

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The assessment of the environmental impacts of chemicals in technical applications is increasingly important as environmentally less burdening substitutes for problematic ones are to be phased-out by law. Life cycle assessment is an appropriate but time-consuming method towards this goal. The intention of this paper is to test if the less laborious mathematical model METEOR (**M**ETHOD of **E**valuation by **O**RDer theory) based on substance-intrinsic properties comes to similar results. In the present work, the environmental impact of eleven refrigerants is studied in their application to mobile air-conditioning systems in passenger cars. The results of two life cycle assessment methods (Eco-Indicator 99, CML02) are compared with those of METEOR. Refrigerants included in this study comprise two hydrocarbons, carbon dioxide, five hydrofluoroethers, one hydrochlorofluorocarbon, and two hydrofluorocarbons, one of which is 1,1,1,2-tetrafluoroethane (R134a), a refrigerant which must be phased out by 2017. The hydrocarbons, carbon dioxide and the other hydrofluorocarbon turn out to be the best alternatives for R134a. METEOR is less subjective than life cycle assessment methods, but the results are not comparable. METEOR is suitable for pre-selection, to keep the number of substances small for which LCA needs to be conducted.

Keywords: life cycle assessment, METEOR, refrigerants, air-conditioning, environmental impact

1. Introduction

Refrigeration is a technical capability which is centrally important for maintaining our present life style. Refrigeration systems are indispensable for safe food transport and distribution, for maintenance of medical and scientific installations, and for comfort of human beings working and living in tropical and

sub-tropical environments. Basically, cooling systems are energy-disproportioning devices which allow to reduce the internal energy of confined spaces at the expense of an overall increase in entropy. Thus, the main environmental impacts are associated with the facts that proportionally more energy is consumed than is transported, depending upon the substances used as refrigerants, and that such substances may have various impacts on the environment associated with their production, unavoidable losses, release and emissions, and their final disposal.

Mobile air-conditioning (A/C) systems of passenger cars were taken as an example of the refrigeration process. The common R134a passenger car A/C system consists of compressor, condenser, accumulator, expansion device, evaporator, and tubes (Fig. 1). All components are connected within a closed cycle. The A/C units extract heat from the vehicle interior and channel it outside. Usually the A/C works with the compression technology. As soon as the A/C unit is switched on at running state of the motor the compressor sucks on the cold and gaseous refrigerant from the accumulator. The refrigerant is condensed, whereby it is heated, and pressed in the condenser.

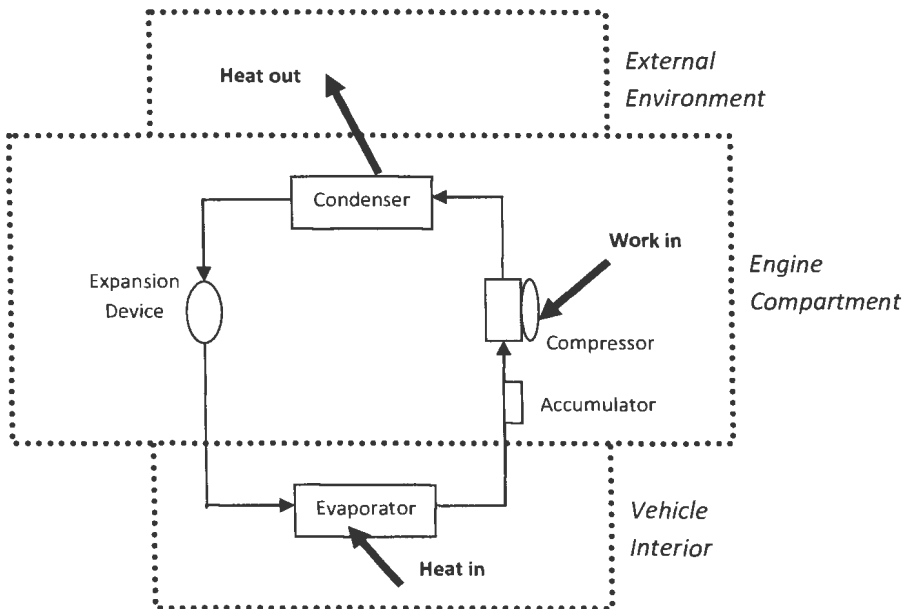


Figure 1. Schematic of the direct expansion R134a A/C system

The air stream of the moving car or from an extra ventilation system cools the condensed, hot gas. As soon as the pressure dependent dew point is reached the refrigerant condenses. The high pressure, liquid refrigerant streams through the expansion device and is being injected into the evaporator. Here it releases tension and evaporates. The required evaporation heat is taken from the air stream that streams around the evaporation fins. In the accumulator it is collected, cleaned and dried. The main leakage points are the shaft sealing of the compressor, the tube system and the gaskets (Schwaab et al., 2004).

In Germany, the A/C sector is with an annual market growth of 4 % the largest in the field of refrigeration and air-conditioning. According to the German Federal Environmental Agency (Schwarz, 2005), A/C systems in passenger cars were the main emission source of fluorinated greenhouse gases in 2002, followed by commercial and industrial refrigeration. As the percentage of passenger cars equipped with A/C systems will reach saturation at 95 % (Schwarz, 2005), this sector is of great interest regarding reductions in ozone depleting substances (ODS), direct greenhouse gas emissions, and in indirect greenhouse gas emissions from energy consumption. In almost all A/C systems in passenger cars, the refrigerant presently used is 1,1,1,2-tetrafluoroethane (R134a) which must be replaced in 2017 according to the Kyoto Protocol. This leads to the necessity of finding the energetically, environmentally and eco/toxicologically least burdening replacements.

In the present study, a life cycle assessment (LCA) referring to A/C systems in passenger cars is conducted for eleven refrigerants, i.e. dichloromethane (R30), 1,1,1,2-tetrafluoroethane (R134a), 1,1-difluoroethane (R152a), propane (R290), isobutane (R600a), carbon dioxide (R744), pentafluorodimethyl ether (E125), 1,1,1',1'-tetrafluorodimethyl ether (E134), heptafluoropropyl methyl ether (E7000), nonafluorobutyl methyl ether (E7100), and nonafluorobutyl ethyl ether (E7200). Two common assessment methods (Eco-Indicator 99, Dutch Handbook method) were applied to the life cycle inventory. The results were compared with those of METEOR (METHod of Evaluation by ORDER theory) considering six refrigerant-intrinsic properties.

2. Scope definition of LCA

The function of the studied A/C system is to keep the passenger compartment at a comfortable temperature of 20 °C, based upon vapour compression and liquid evaporation. The above mentioned refrigerants are applied to the A/C system. The chemical formulae and names of those refrigerants are listed in Table 1.

Table 1. Refrigerants applied to A/C system in the present study

Refrigerant	Chemical formula	Chemical name
R30	CH ₂ Cl ₂	Dichloromethane
R134a	C ₂ H ₂ F ₄	1,1,1,2-Tetrafluoroethane
R152a	C ₂ H ₄ F ₂	1,1-Difluoroethane
R290	C ₃ H ₈	Propane
R600a	C ₄ H ₁₀	Isobutane
R744	CO ₂	Carbon dioxide
E125	CF ₃ -O-CHF ₂	Pentafluorodimethyl ether
E134	CHF ₂ -O-CHF ₂	1,1,1',1'-Tetrafluorodimethyl ether
E7000	C ₃ F ₇ -O-CH ₃	Heptafluoropropyl methyl ether
E7100	C ₄ F ₉ -O-CH ₃	Nonafluorobutyl methyl ether
E7200	C ₄ F ₉ -O-C ₂ H ₅	Nonafluorobutyl ethyl ether

In general, the scope of an LCA comprises input and output of production, operation (including servicing/refilling), and disposal phase (Fig. 2). Inventory data are literature values.

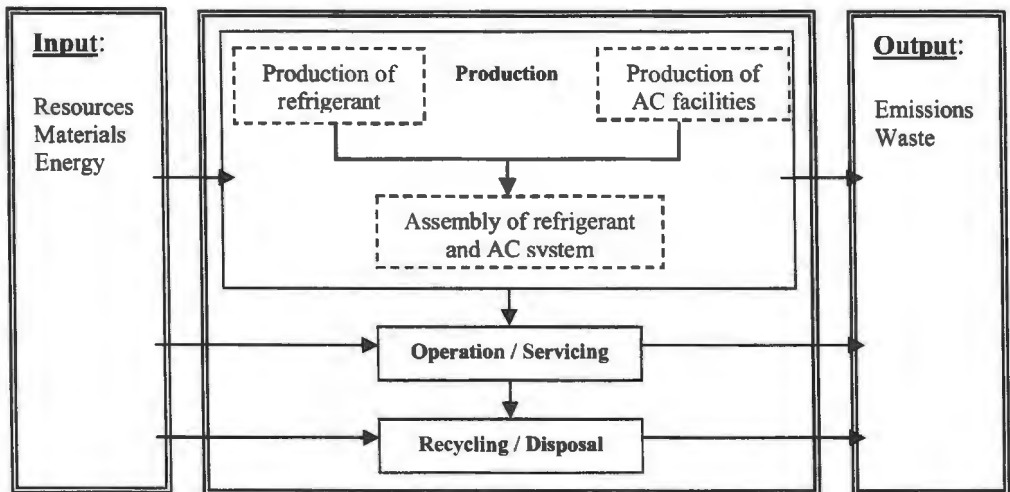


Figure 2. Schematic of the life cycle of A/C system

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Worst-case, best-case, and average scenarios were defined taking different operation times, leakage rates, and servicing intervals into account. The A/C system is operated with respect to climate conditions of a cold (best-case scenario), moderate (average scenario), and warm (worst-case scenario) European country within a 10 years lifetime of the passenger car. The refrigerant leakage within the life cycle is set to be independent of the refrigerant but dependent on the different life stages. Three emission scenarios are defined and the leakage rates of the different life phases were estimated (Table 2). During the usage phase, the A/C system is serviced and refilled four times under worst-case, twice under average, and zero times under best-case conditions, resulting in an emission of 100 g refrigerant per servicing. The A/C system is emptied and then refilled with new refrigerant.

Table 2: Direct emission scenarios within the life cycle of an A/C system, wc – worst-case scenario, a – average scenario, bc – best-case scenario

Life cycle phase	Place of emission	Direct refrigerant emissions scenario [% refrigerant charge]		
		wc	a	bc
Manu-facture	Production of refrigerant	1	0.5	0.1
	Loading of tanks/bottles	5	2	1
	Charging of A/C system	5	2	0.5
Usage	Regular emissions	100	77	30
	Irregular emissions	100	33	0
Disposal	Disposal vehicle	100	50	50
Total		311 *)	164.5 *)	81.6

*) Plus 0.4 kg additional refrigerant for four servicing events during lifetime (Schwarz and Harnisch, 2003), **) Plus 0.2 kg additional refrigerant for two servicing events during lifetime (Schwarz and Harnisch, 2003)

3. Assessment methods

As mentioned in the introduction, two different assessment methods were applied to the results of the life cycle inventory in order to find as replacement for R134a the refrigerant least harmful to the environment. The results of the two

methods, namely the Dutch Handbook method (CML02) and Eco-Indicator 99 (EI99), will be compared with those based on METEOR.

3.1. CML02

In this study, ten impact categories were considered following the mid-point assessment method CML02 (Guinée et al., 2001) to evaluate the environmental impact of certain refrigerants in A/C systems in passenger cars: Demand of non-renewable primary energy (PE), Depletion of abiotic resources (excluding primary energy sources) (ADP), Climate change (GWP), Stratospheric ozone depletion (ODP), Human toxicity (HTP), Photo-oxidant formation (POCP), Acidification (AP), Eutrophication (EP), Fresh water aquatic toxicity (FAETP), and Terrestrial ecotoxicity (TETP). The impact factors of the different categories are multiplied with the respective emissions, which contribute to the specific category, arising during the life cycle (Guinée et al., 2001).

3.2. EI99

The EI99 is a damage-oriented impact assessment method for LCA which constitutes the basis for the calculation of eco-indicator scores for materials and processes (Goedkoop and Spriensma, 2001). This end-point method is divided into different steps such as fate, exposure, effect, and damage analysis which are combined in the damage factors which are multiplied with the respective emissions arising during the life cycle. The method accounts for the contribution of emitting substances to three damage categories which are Human Health, Ecosystem Quality, and Resources. The method includes normalisation and weighting of the three damage categories resulting in one index call Eco-Indicator 99.

4. METEOR

METEOR (**M**ETHOD of **E**valuation by **O**Rder theory), a mathematical method for assessing descriptor prioritisation and its effect on the ranking of substances, was used for evaluating the environmental impact of refrigerants. This method is based on discrete mathematics and directed graphs (Brüggemann and Bartel, 1999; Brüggemann and Münzer, 1993; Restrepo et al., 2008).

In the present work, the idea of METEOR was applied considering six refrigerant-intrinsic properties (critical temperature, heat capacity of vapour, global warming potential, ozone depletion potential, octanol-water partition coefficient,

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and toxicological exposure limits) for a selection of seven of the mentioned refrigerants (E7200, R134a, R152a, R290, R30, R600a, R744).

The first step using METEOR was to normalize the data values of the refrigerant-intrinsic properties to a [0,1]-scale using equation (1) and to reorient the normalised values using equation (2) where necessary, so that high values are associated with a negative environmental impact (Table 3).

$$q_i(x) = \frac{q_i'(x) - \min q_i'}{\max q_i' - \min q_i'} \quad (1)$$

$$q_{i,re}(x) = [q_i(x) \cdot (-1)] + 1. \quad (2)$$

For equation (1) and (2): $q_i'(x)$ is the value of property i for refrigerant x and $\min q_i'$ and $\max q_i'$ are the minimum and maximum values, $q_{i,re}(x)$ is the reoriented, normalised valued of $q_i'(x)$.

Table 3. Normalised and reoriented data values for certain refrigerants

Refrigerant	t_c	c_p	GWP	ODP	c_{ow}	expos
R134a	0.6596	0.1690	0.3205	0.0003	0.5844	0.8081
R152a	0.6019	0.4733	0.0277	0	0.5086	0.8081
R290	0.6805	0.3801	0.0045	0	0.9022	0.5051
R600a	0.4951	0	0.0045	0	1	0.8485
R744	1	1	0.0002	0	0	0
E7200	0.1359	0.3476	0.0136	0	0.5826	0.9697
R30	0	0.7814	0.0023	0.01	0.6308	1

The second step was to aggregate two properties each time following equations (3) to (5). The critical temperature (t_c) was aggregated with heat capacity of vapour (c_p) resulting in ϕ_1 , global warming potential (GWP) with ozone depletion potential (ODP) resulting in ϕ_2 , and octanol-water partition coefficient (c_{ow}) with exposure limit ($expos$) resulting in ϕ_3 :

$$\phi_1(x) = g \cdot t_c(x) + (1 - g) \cdot c_p(x) \quad (3)$$

$$\varphi 2(x) = g \cdot GWP(x) + (1 - g) \cdot ODP(x) \tag{4}$$

$$\varphi 3(x) = g \cdot c_{ow}(x) + (1 - g) \cdot expos(x) \tag{5}$$

where g and $(1-g)$ are the selected weights for the properties. The sum of the weights must be equal to 1. An important value of g is achieved when $\varphi(x) = \varphi(y)$. This particular g -value is called “crucial” g -value for the pair $\{x,y\}$ (Restrepo et al., 2008).

Each aggregation delivers a set of crucial g -values (Fig. 4) that separates the range of g from 0 to 1 into a number of different stability fields. For further calculations, from each aggregation the stability fields were chosen with a range of g -values which are equal or greater than half of the range of greatest stability field of the respective aggregation (Fig. 3). Each of those stability fields has its characteristic linear order (Fig. 4). The other stability fields were pooled to so called hot spots, marked with grey bars (Fig. 3), and not further analysed.

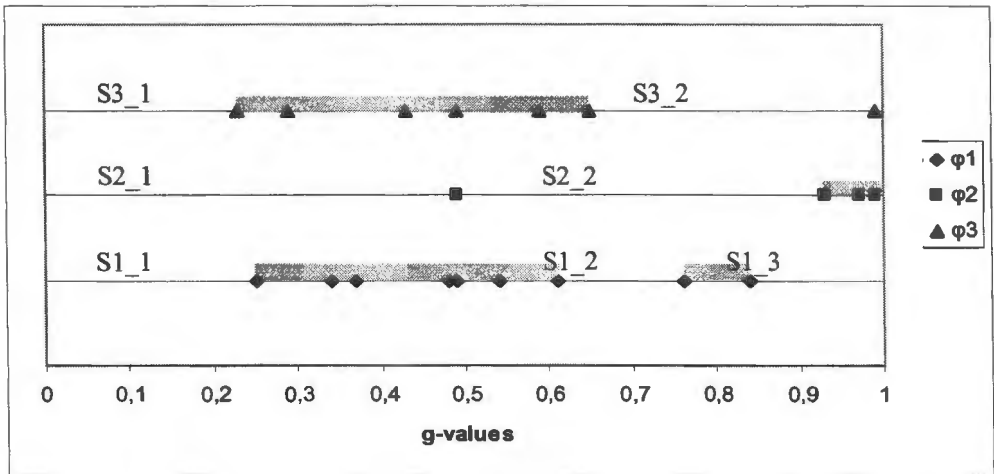


Figure 3. Crucial g -values of the aggregations $\varphi 1$ (t_c, c_p), $\varphi 2$ (GWP, ODP), and $\varphi 3$ ($c_{ow}, expos$), marked with grey bars are the stability fields that are combined to hot spots, further analysed stability fields are labelled

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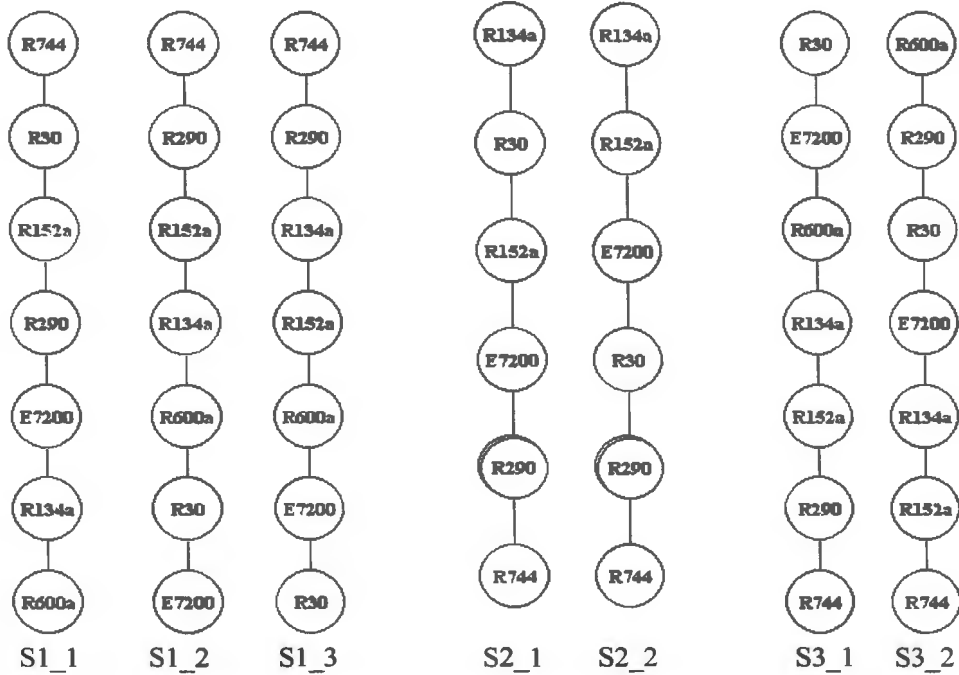


Figure 4. Characteristic linear extensions of the selected stability fields, in S2_1 and S2_2 the refrigerants R290 and R600a have the same rank, only R290 is displayed

The next step was to draw Hasse diagrams using a combination of three linear extensions, one from each aggregation (Table 3).

Table 3. Set of linear extensions used for drawing Hasse diagrams

Hasse diagram No.	$\phi 1$	$\phi 2$	$\phi 3$	Range of g-values (%)					
				t_c	c_p	GWP	ODP	c_{ow}	expos
1	S1_1	S2_1	S3_1	0-25	75-100	0-49	51-100	0-23	77-100
2	S1_1	S2_1	S3_2	0-25	75-100	0-49	51-100	65-99	1-35
3	S1_1	S2_2	S3_1	0-25	75-100	49-93	7-51	0-23	77-100

4	S1_1	S2_2	S3_2	0-25	75-100	49-93	7-51	65-99	1-35
5	S1_2	S2_1	S3_1	61-76	24-39	0-49	51-100	0-23	77-100
6	S1_2	S2_1	S3_2	61-76	24-39	0-49	51-100	65-99	1-35
7	S1_2	S2_2	S3_1	61-76	24-39	49-93	7-51	0-23	77-100
8	S1_2	S2_2	S3_2	61-76	24-39	49-93	7-51	65-99	1-35
9	S1_3	S2_1	S3_1	84-100	0-16	0-49	51-100	0-23	77-100
10	S1_3	S2_1	S3_2	84-100	0-16	0-49	51-100	65-99	1-35
11	S1_3	S2_2	S3_1	84-100	0-16	49-93	7-51	0-23	77-100
12	S1_3	S2_2	S3_2	84-100	0-16	49-93	7-51	65-99	1-35

5. Results

5.1. METEOR

In total, twelve Hasse diagrams were drawn of which six were different (Fig. 5). When a low weight was given to t_c (0-25 %) and GWP (0-49 %) and respective a high weight to c_p (75-100 %) and ODP (51-100 %), R30 has greater environmental impact than R152a and E7200; R134a and R744 are incomparable to the other refrigerants (HD 1, HD 2).

In case of an additionally low weight on c_{ow} (0-23 %) and therefore a high weight on $expos$ (77-100 %), R290 and R600a have lower impacts than R30, R152a, and E7200 (HD 1). However, when a high weight is placed on c_{ow} (65-99 %) and a low on $expos$ (1-35 %) R290 and R600a become incomparable to the other refrigerants (HD 2).

When using low weight on t_c (0-25 %) and c_{ow} (0-23 %) but high weight on GWP (49-93 %), R290 is dominated by R152a and R30, and R600a is dominated by R30 and E7200 (HD 3).

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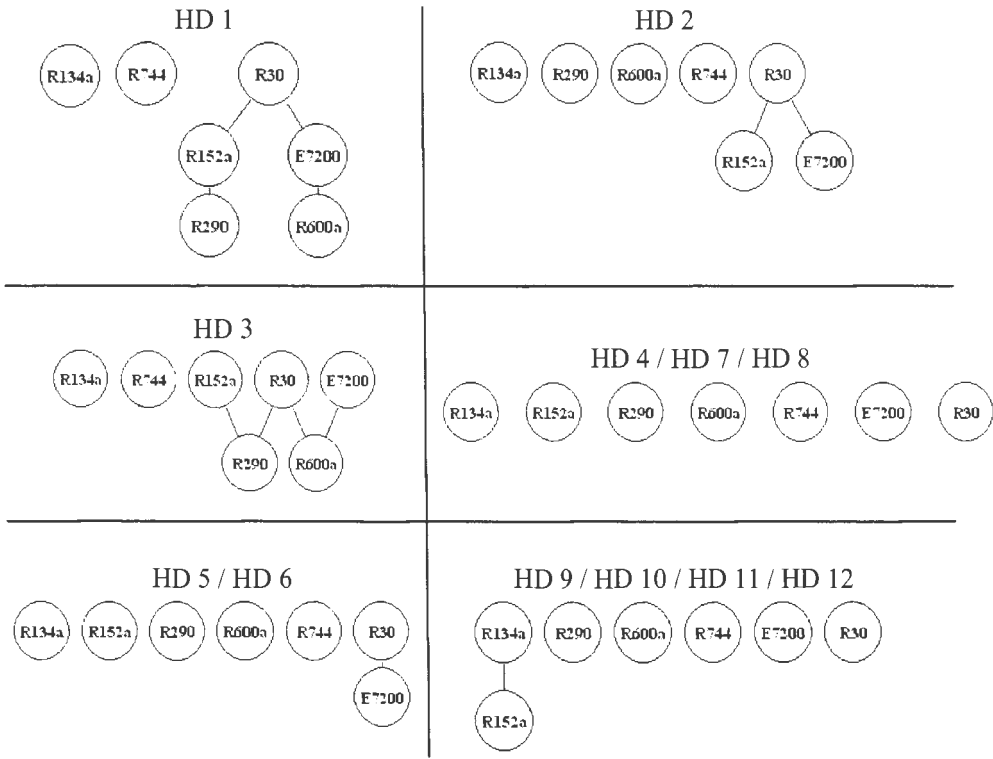


Figure 5. Hasse diagrams using different combinations of aggregated functions (Table 3)

By prioritising t_c (61-76 %) and ODP (51-100 %), R30 is having a higher environmental impact than E7200 regardless of the weighting of c_{ow} and $expos$ (HD 5 and HD 6).

Only by applying high weight to t_c (84-100 %), R134a becomes comparable to another refrigerant; it is then dominating R152a (HD 9 to HD 12). Overall, many incomparabilities occur using those refrigerant-intrinsic properties.

5.2. LCA

The rankings from the three different scenarios (Table 4) are employed as parameters for drawing two Hasse diagrams, one for each assessment method.

Table 4. Ranks of refrigerants derived from EI99 and CML02 assessment method, CML02_{average} rank was calculated from ranks of the 10 impact categories of CML02, bc – best-case scenario, a – average scenario, wc – worst-case scenario

	EI99			CML02 _{average}		
	bc	a	wc	bc	a	wc
E125	7.0	8.0	6.0	4.8	4.7	4.7
E134	11.0	11.0	11.0	7.9	7.9	7.8
E7000	9.0	6.0	7.0	5.7	4.5	4.4
E7100	6.0	7.0	8.0	4.6	5.3	5.4
E7200	5.0	5.0	5.0	5.3	4.3	4.3
R134a	10.0	9.0	9.0	6.4	7.6	7.6
R152a	4.0	3.0	3.0	5.5	5.2	5.2
R290	1.0	1.0	1.0	3.9	3.9	3.9
R600a	2.0	2.0	2.0	4.4	4.4	4.4
R744	3.0	4.0	4.0	5.1	5.1	5.1
R30	8.0	10.0	10.0	6.5	7.2	7.3

EI99 entails a Hasse diagram that places E134 as maximum element and R290 as minimum element considering all three scenarios at the same time (Fig. 6, left). R134a has higher environmental impact than E7000, E7100, E7200, E125, R152a, R744, R600a, and R290. R134a is incomparable to R30. The HCs, R744, and R152a have a smaller environmental impact than the HFEs, R30, and R134a. This confirms them as possible replacements of R134a.

The CML02 assessment method comprises 10 independent impact categories, each with its own ranking of the refrigerants. For each scenario, an average ranking (CML02_{average}) was calculated weighting the ranks of every refrigerant in each impact category equally (Table 4). The Hasse diagram (Fig. 6, right) using CML02_{average} places E134 as the maximum element. R134a and R30 are ranked lower than E134 but higher than the other refrigerants. R290 is ranked as minimum element, similar to the EI99 method. However, E125 and R744 are not comparable to the three refrigerants of the E7000-series. Contrary to EI99, E125 is ranked lower than R152a and R744. R290 and R600a are less problematic than R152a and R744, as was the case for EI99.

Comparing the METEOR results with those from LCA, some agreements were found. For example, in both LCA methods and in some METEOR Hasse diagrams (HD 1, HD 2, HD 3) the domination of R30 over R152a, E7200, R600a, and/or

R290 was observed. The same holds for the domination of R152a over R290 (HD 1, HD 3). When R134a and R152a were comparable in the METEOR Hasse diagrams (HD 9 to HD 12), they match the order relation of the LCA results; R152a is placed lower than R134a. On the other hand, CML02 does not show a dominance of E7200 over R600a.

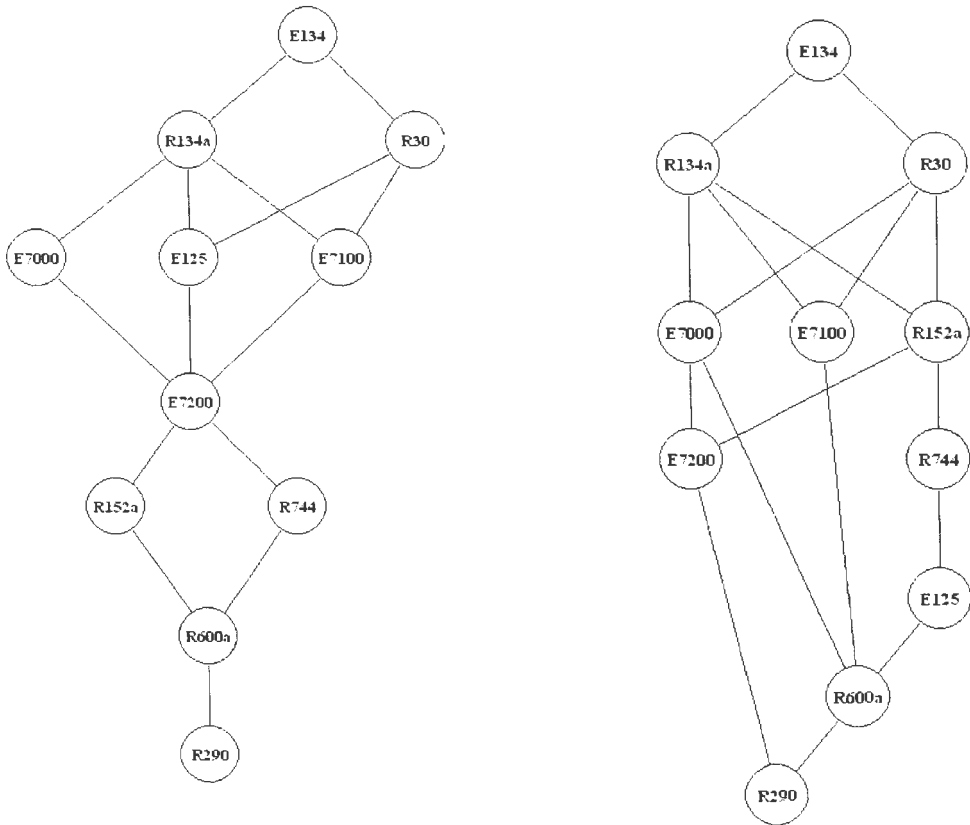


Figure 6. Hasse diagrams using the worst-case, best-case, and average results as parameters for EI99 (left) and CML02_{average} (right)

Because of the many incomparabilities within the METEOR Hasse diagrams HD 1 to HD 12, a comparison with LCA diagrams is not easy. Therefore, a new set of METEOR Hasse diagrams was drawn using only two linear orders at one time. Exemplary combinations of linear orders are shown in Table 5. The corresponding Hasse diagrams are shown in Fig. 7.

Table 5. Second set of linear extensions used for drawing Hasse diagrams using only two aggregations as parameters

Hasse diagram No.	$\phi 1$	$\phi 2$	$\phi 3$	Range of g-values (%)					
				t_c	c_p	GWP	ODP	c_{ow}	expos
13	S1_1	S2_1		0-25	75-100	0-49	51-100		
14	S1_1		S3_1	0-25	75-100			0-23	77-100
15	S1_3	S2_1		84-100	0-16	0-49	51-100		
16		S2_1	S3_1			0-49	51-100	0-23	77-100
17		S2_1	S3_2			0-49	51-100	65-99	1-35
18		S2_2	S3_1			49-93	7-51	0-23	77-100

Whenever a linear order of $\phi 1$ (t_c , c_p) is taken as one parameter for drawing a Hasse diagram (HD 13, HD 14, HD 15), R744 is incomparable to the other refrigerants. This is due to the different ranking of R744 in $\phi 1$ (rank = 7) and the other two aggregations $\phi 2$ and $\phi 3$ (rank = 1). R744 can therefore not be compared with the other refrigerants considering the substance-intrinsic properties t_c and c_p . This is of disadvantage because especially the thermodynamic properties describe the technical efficiency of a refrigerant.

The higher ranking of R744 compared to the HCs (R290, R600a) in EI99 and CML02_{average} is not reflected in Hasse diagrams HD 16, HD 17, and HD 18 of METEOR. However, the dominance of E7200, R134a, and R30 over R744 is reflected. Moreover, Hasse diagrams HD 16, HD 17, and HD 18 show also the incomparability of R134a with R30, the dominance of R600a over R290, and the higher ranking of R134a compared with R152a as do the LCA Hasse diagrams.

HD 16 and HD 18 reflect the LCA diagrams in additional points. Namely the higher ranking of R134a and R152a compared with R290. From those chosen secondary set of Hasse diagrams, HD 16 and HD 18 match those from LCA best. This means that with a high weight placed on ODP (51-100 %) or GWP (49-93 %) and simultaneously on expos (77-100 %) similar results to LCA are gained.

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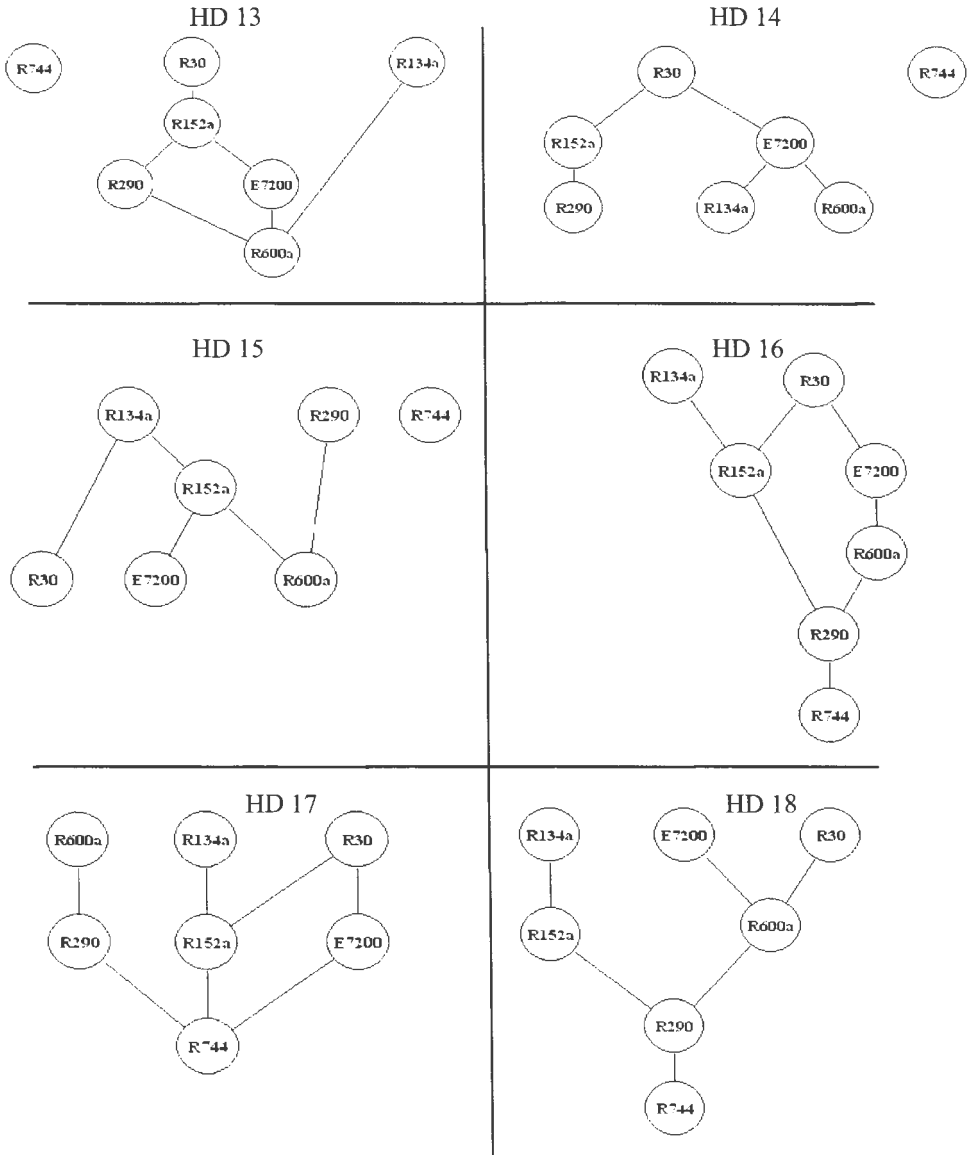


Figure 7. Second set of Hasse diagrams using different combinations of aggregated functions (Table 5)

6. Conclusions and outlook

The idea of the present study was to check if methods using discrete mathematics and directed graphs based on substance-intrinsic properties give the same or similar evaluation of the environmental behaviour of refrigerants as can be derived by LCA.

The set of criteria for which Hasse diagrams for METEOR and the LCA methods were drawn are generally different. METEOR consisted at the first attempt of three aggregated parameters each derived from the aggregation of two refrigerant intrinsic properties. In the second attempt, only two aggregated parameters were used to draw Hasse diagrams. Each LCA method created Hasse diagrams using three rankings from different life cycle scenarios respectively. But regardless of the different sets of criteria, the ranking of refrigerants relatively to each other was thought to can be made visual using directed graphs.

In conclusion, it can be stated that the environmental impact assessment by means of METEOR can give a preliminary estimate for some substances and is less subjective than LCA methods. The objectivity of METEOR is bound to the choice of weights. Because of the selection of stability fields made in this study, it is not possible to place an equal weight on all six refrigerant intrinsic properties. One has to place a higher weight on either t_c or c_p . By giving a higher weight to t_c , the Hasse diagram is influenced considerably as this set of weights results finally in the dominance of R134a over R152a. Whereas, in all other combinations of weights, R134a is not comparable to any other refrigerant.

By reducing the number of parameters to two, METEOR shows more comparable results to LCA diagrams. In supplementary studies, φ_1 , φ_2 , and φ_3 will be further aggregated. Finally, linear rankings will be calculated with specific sets of weighted refrigerant-intrinsic properties. Those might be used to make a pre-selection of the least environmental harmful substances, an elimination of the most problematic substances, and to conduct a complete LCA on pre-selected chemicals. METEOR cannot completely substitute the more time-consuming process of LCA, which also takes the consequences of the technical application of the refrigerants into account.

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