Emissions and Reduction Potentials of Hydrofluorocarbons, Perfluorocarbons and Sulphur Hexafluoride in Germany

by

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16.	Abstract					
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Abstract

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Emissions and Reduction Potentials of Hydrofluorocarbons, Perfluorocarbons and Sulphur Hexafluoride in Germany

Introduction

The Protocol adopted on 10 December 1997 in Kyoto at the 3rd Conference of the Parties to the UN Framework Convention on Climate Change commits industrialized nations for the first time in legally binding form to reduce greenhouse gas emissions. Worldwide, these are to be reduced by 2008/2012 by 5.2% from 1990 levels. A number of industrialized countries, including the EU, have committed themselves to larger reductions. Under a decision taken by the EU Council of Ministers in June 1998, Germany has an above-average commitment within the burden-sharing scheme agreed within the EU. Germany has committed itself to reducing total greenhouse gas emissions by 21% by 2008/2012.

The Kyoto Protocol covers not only CO_2 , methane and nitrous oxide, but also fluorinated gases: hydrofluorocarbons (HFCs), perfluorocarbons (PFCs) and sulphur hexafluoride (SF₆). The 5.2% (worldwide) or 21% (German) reduction target refers not to each individual one of these six gases, but to their total "basket" expressed in CO_2 equivalent emissions. The general baseline year is 1990; for the three fluorinated gases, 1995 can also be chosen.

The propensity of fluorinated compounds (FCs) to cause a radiative forcing results from their long atmospheric lifetimes of several decades (HFCs) to centuries (PFCs, SF₆), in conjunction with high specific absorption in the infrared spectral region of relevance to global warming. FCs are by far the most potent greenhouse gases in the atmosphere. The Global Warming Potential (GWP) - a comparative index in relation to CO_2 as reference substance - of HFCs and PFCs is in the range of 1000 to 10,000. That of SF₆ even figures 23,900.

The global warming relevance of FCs has only been studied in more detail in recent years. It became clear that while their emission sources are understood in a general way, too little concrete knowledge of the quantitative contributions of the individual sources is available. Studies have therefore been conducted in many countries with the aim of identifying the applications of fluorinated greenhouse gases, calculating their present and future consumption and emission levels, and appraising emission abatement options.

It is in this context that the present study on emissions of global warming FCs in Germany was conducted. It builds upon a first study of HFCs, PFCs and SF₆ conducted in 1996 on behalf of the German Federal Environmental Agency (Umweltbundesamt, UBA).¹

¹ Schwarz, W./Leisewitz, A. (Öko-Recherche): Aktuelle und künftige Emissionen treibhauswirksamer fluorierter Verbindungen in Deutschland, for Umweltbundesamt, Berlin, December 1996.

Summary

On the basis of the historical consumption and emission levels of the three fluorinated greenhouse gases - HFCs, PFCs and SF₆ - in Germany from 1995 to 1997, emissions forecasts until 2010 are elaborated for each individual application sector, with and without additional abatement measures beyond business-as-usual.

A business-as-usual scenario assumes that present usage trends, including emission control measures already instituted today, continue, and extrapolates these to the year 2010. A reduction scenario assumes that existing technology potentials for abating or substituting emissions are exploited in each individual emission sector. In the reduction scenario, only emission reduction measures are included whose ecological effect is not neutralized by disadvantages elsewhere.

Neither business-as-usual nor the reduction scenario yield a stabilization of FC emissions. However, their rise by 150% over the 1995-2010 period in the business-as-usual scenario can be limited to a 34% rise in the reduction scenario.

1 2010 Emissions of HFCs, PFCs and SF₆ in the Business-as-Usual Scenario

In the business-as-usual scenario, total HFC, PFC and SF₆ emissions grow over the 1995-2010 period from 11.1 to 27.4 million tonnes CO_2 equivalent, or by 146% (see Table I).

As to be expected, HFC emissions grow most, as these have only been produced specifically as substitutes for CFCs and HCFCs since 1990. HFC emissions grow over the 1995-2010 period from 3.1 million t CO_2 equiv. to 19.8 million t, or by a factor of about 6. Overall FC emissions growth is mainly attributable to HFCs (cf. Table I and Diagram I).

Table I: 1995-2010 emissions of the three fluorinated greenhouse gases							
in the	in the business-as-usual scenario [million t CO ₂ equivalent]						
	1995	2000	2005	2010	1995-2010		
1. HFCs	3.13	8.35	14.98	19.84	+ 534%		
2. SF ₆	6.22	3.69	4.01	5.01	- 19%		
3. PFCs	1.76	1.83	2.03	2.52	+ 43%		
Total	Total 2116 2199 21.02 27.37 + 146%						

 SF_6 emissions drop over the same period from 6.2 to 5.0 million t CO_2 equivalent, or by 19%. PFC emissions, in contrast, rise by 43% from 1.8 to 2.5 million t CO_2 equivalent.

Ranking emissions sources in the business-as-usual scenario

With business-as-usual, it is to be expected that in the year 2010 86% of FC emissions, amounting to 23.6 million t CO_2 equivalent, will be attributable to the seven largest of the eighteen FC application sectors. These seven include four HFC applications, two PFC sources and one SF₆ application. The following pie-charts show the shares of the various applications in the total CO_2 equivalent emissions of the three FCs.



The most weighty sectors (cf. Table III), in terms of their contributions to total FC emissions, are, in the following order:

1. Refrigeration and stationary air-conditioning (HFCs)	7.7 mill. t CO ₂ equiv.
2. Mobile air-conditioning (HFCs)	4.7 mill. t CO ₂ equiv.
3. Blowing extruded polystyrene (HFCs)	3.0 mill. t CO ₂ equiv.
4. Soundproof glazing (SF ₆)	3.0 mill. t CO ₂ equiv.
5. One-component polyurethane foam (HFCs)	2.7 mill. t CO_2 equiv.
6. Semiconductor manufacturing (PFCs)	1.4 mill. t CO ₂ equiv.
7. Aluminium smelting (PFCs)	1.1 mill. t CO ₂ equiv.

2 2010 Emissions of HFCs, PFCs and SF₆ in the Reduction Scenario

In the reduction scenario, not only emissions of SF_6 drop by 2010 from the 1995 baseline, but also those of PFCs. SF_6 emissions halve from 6.2 to less than 3.0 million t CO_2 equivalent (Table II). PFC emissions drop from 1.8 to 1.1 million t CO_2 equivalent, or by 35%, instead of rising - as in the business-as-usual scenario - by 43%.

Table II: 1995-2010 emissions of the three fluorinated greenhouse gases							
i	in the reduction scenario [million t CO ₂ equivalent]						
	1995 2000 2005 2010 1995-2010						
1. HFCs	3.13	7.59	10.67	10.77	+ 244%		
2. SF ₆	6.22	3.53	2.66	2.95	- 53%		
3. PFCs	1.76	1.76	1.44	1.14	- 35%		
Total	2116	2199	14.77	14.86	+ 34%		

The growth of HFC emissions is attenuated. These rise from 3.1 to 10.8 million t CO_2 equivalent (Table II), i.e. by 244% instead of by 534% as in the business-as-usual scenario. However, this growth is still enough to make total FC emissions rise from 1995 to 2010 by 34%, from 11.1 to 14.9 million t CO_2 equivalent (Table II).

The reduction potential in the year 2010 of all measures proposed totals about 12.5 million t CO_2 equivalent, or 46%.

Sector-by-sector reduction potentials

Beyond the absolute quantities of individual emission sectors, it is essential to environmental policy to know which sectors offer the greatest opportunities to abate radiative forcing emissions. In general, the largest emissions sources also offer the largest absolute reduction potential (cf. Table III).

Five of the eighteen emission sectors offer a reduction potential of more than 1 million t CO_2 equivalent in the 1995-2010 period: the three HFC applications <u>refrigeration and</u> <u>stationary air-conditioning</u>, <u>one-component PU foam</u> and <u>XPS foam blowing</u>, the SF₆ application <u>soundproof glazing</u> and the PFC application <u>semiconductor manufacturing</u> (cf. Table III, right column).

- The largest reduction potential is in <u>refrigeration and stationary air-conditioning</u>, which is also the largest source of emissions in the business-as-usual scenario. In the reduction scenario, emissions in 2010 only figure 4.1 instead of 7.7 million t CO₂ equivalent 3.6 million t less than in the business-as-usual scenario. This reduction is the outcome of general mandatory maintenance for refrigeration and air-conditioning systems containing more than 1 kg refrigerant. This alone cuts total business-as-usual emissions of HFCs, PFCs and SF₆ (27.4 million t CO₂ equivalent) by 13%.
- Total emissions are further reduced by 9% by means of partial HFC substitution in propellants for <u>one-component PU foam</u>, namely 95% substitution by simple hydrocarbons. This measure has the potential to slash business-as-usual HFC emissions (2.7 million t CO₂ equivalent) in 2010 in this sector to 0.2 million t.
- The third largest reduction effect compared to the business-as-usual scenario is also provided by HFC substitution. If in about 40% of <u>XPS foam panels</u> HFCs are replaced as blowing agent by CO₂ plus ethanol, then emissions from this application are cut from 3 million t CO₂ equivalent to 1.7 million t. This measure alone would reduce total FC emissions in the year 2010 by 1.3 million t CO₂ equivalent, or 5%.
- The fourth largest reduction potential is available in the <u>semiconductor industry</u>, if from 2000 onwards process chamber cleaning converts throughout to a new technology (upstream NF₃ dissociation in microwave plasma). PFC emissions then drop from 1.4 million t CO₂ equivalent in the business-as-usual scenario to less than 0.2 million t CO₂ equivalent in the reduction scenario. This measure reduces total FC emissions by more than 4%.
- A reduction potential of 1 million t CO₂ equivalent could be tapped if SF₆ were no longer used in new <u>soundproof glazing</u>. Of the 3 million t CO₂ equivalent emitted from this sector in the year 2010 in the business-as-usual scenario, 2 million t would remain in the reduction scenario due to decommissioning losses. The high filling losses would cease.

Table III: Application sectors of fluorinated greenhouse gases,							
in the order of emission	in the order of emissions in 2010 [million t CO ₂ equivalent] in both scenarios						
	Emissions [mill.	t CO ₂ equivalent]	Reduction				
Application			potential*				
	in b-a-u scenario	in reduction scenario					
1. Refrig. & stationary A/C	7.66	4.11	3.55				
2. Mobile A/C	4.69	3.86	0.83				
3. XPS foam	3.04	1.73	1.31				
4. Soundproof glazing	3.02	2.01	1.01				
5. One-comp. PU foam	2.74	0.19	2.55				
6. Semicond. (PFCs)	1.45	0.19	1.26				
7. Aluminium smelting	1.05	0.92	0.13				
8. Car tires	0.72	0.00	0.72				
9. Metered-dose inhalers	0.68	0.17	0.51				
10. PU foam products	0.68	0.45	0.23				
11. Others	1.70	1.29	0.40				
Total	Total 565 403 458						

* Difference between business-as-usual and reduction scenarios in million t CO₂ equivalent.



Diagram I: 1995-2010 emissions of the three gases in business-as-usual Scenario I [million t CO_2 equivalent]. The steep rise in emissions until 2010 to more than 27 million t CO_2 equivalent is almost exclusively attributable to HFCs (dark upper area). The sub-total of SF₆ and PFC emissions (lower areas) drops until 2001, but then rises again until 2010.



Diagram II: 1995-2010 emissions of the three fluorinated greenhouse gases in reduction Scenario II [million t CO_2 equivalent]. Both SF_6 and PFC emissions (lower areas) drop over the 1995-2010 period. However, what is decisive for the reduction potential (white uppermost area) from the year 2000 onwards is the moderate rise in HFC emissions compared to Scenario I.

 The sectors offering reduction potentials of more than 1 million t CO₂ equivalent effectively also include the second largest emissions source of the business-asusual scenario, <u>mobile air-conditioning</u>. The reduction potential only reaches 0.8 million t CO₂ equivalent by the year 2010, but this is merely because the emissionabating factor, namely refrigerant conversion from HFCs to CO₂, only develops its full effect a few years later.²

The above six measures can tap 84% of the total reduction potential of 12.5 million t CO_2 equivalent (Table III, right column).

3 Conclusions for Emissions Abatement Policies

Where emissions abatement policies need to set priorities, the finding that the bulk of the fluorinated greenhouse gas reduction potential in Germany is concentrated in six application sectors provides guidance. Nonetheless, it should be kept in mind that the other twelve sectors offer a substantial further reduction potential totalling 2 million t CO_2 equivalent.

It is all the more important to keep this in mind in view of the further finding that even if all measures proposed are implemented, emissions will by no means be brought below the 1995 baseline by 2010. Germany has committed itself to <u>reduce</u> by 21%, from the 1995 baseline, the total of all greenhouse gas emissions (fluorinated and nonfluorinated compounds). In contrast, exploiting the full reduction potential set out in this study for the three groups of fluorinated greenhouse gases leads not to a reduction, but to a <u>rise</u> of global warming emissions by 34%. This growth has to be compensated elsewhere (CO_2 , CH_4 , N_2O). It is thus amply clear that no options to reduce global warming emissions should be neglected. On the contrary, efforts are necessary to tap further reduction potentials.

By the measure of the overall emissions of direct greenhouse gases in Germany, the emissions of HFCs, PFCs and SF₆ may appear relatively unimportant. Together, their share in total emissions - expressed in CO_2 equivalents (GWP with 100 year time horizon) - amounted to ca. 1% in 1995. Nonetheless, it would be ecologically inappropriate to neglect fluorinated greenhouse gases. For if no further abatement measures are implemented, their share in total direct greenhouse gas emissions will have risen to some 3% by 2010. A further aspect is that, over the medium term, no reversal of emission trends is in sight for the quantitatively most important group of fluorinated gases, the HFCs.

It must further be noted that the total of global warming emissions is made up of a great array of individual sources, each of which is generally small in itself, but all of which need to be scrutinized for abatement options as a part of the crucial effort to reduce greenhouse gas emissions.

² From 2020 onwards, HFC emissions from mobile air-conditioning could cease entirely, which would mean an emissions reduction of 4.7 million t CO₂ equivalent per annum.

Annex: Supplementary data

Tables IV to VII serve to complete the environmentally relevant emission data for fluorinated greenhouse gases in Germany.

Tables IV and V list gas emissions for the years 1995, 2000, 2005 and 2010 in tonnes per year. Table IV gives the figures for the business-as-usual scenario, Table V for the reduction scenario.

Table IV: 1995-2010 emissions of the three fluorinated greenhouse gases							
in the	in the business-as-usual scenario in tonnes of gas per year						
	1995	2000	2005	2010	1995-2010		
1. HFCs	2135	9298	14361	18825	+ 782%		
2. SF ₆	260	154	168	210	- 20%		
3. PFCs	258	259	181	340	+ 32%		
Total	Total 4648 11711 16715 21385 + 630%						

Table V: 1995-2010 emissions of the three fluorinated greenhouse gases							
i	in the reduction scenario in tonnes of gas per year						
1995 2000 2005 2010 1995-2010							
1. HFCs	2135	7294	9421	9984	+ 368%		
2. SF ₆	260	148	111	124	- 52%		
3. PFCs	258	250	204	165	- 36%		
Total	2653	7692	11741	12283	+ 287%		

Tables VI and VII list the historical emissions of the years 1995 to 1997: Table VI in tonnes of gas and Table VII in million tonnes CO_2 equivalent.

Table VI: 1995-1997 emissions of the three fluorinated greenhouse gases						
in t/a						
1995 1996 1997						
1. HFCs	2135	2435	2884			
2. SF ₆	260	243	238			
3. PFCs	258	265	221			
Total	4648	4939	5340			

Table VII: 1995-1997 emissions of the three fluorinated greenhouse gases in million tonnes CO ₂ equivalent						
1995 1996 1997						
1. HFCs	3.13	2.58	3.45			
2. SF ₆	6.22	5.81	5.70			
3. PFCs	1.76	1.83	1.55			
Total	2116	2226	21760			

I. Hydrofluorocarbons (HFCs)

Overview

Since the late 1990s, hydrofluorocarbons (HFCs) are supplied by the chemical industry as ozone-neutral replacements for CFCs and HCFCs. Their global warming potential (GWP), which is generally much lower than that of CFCs and HCFCs but still considerable, presents the question of the global warming impacts that they generate. In order to assess future HFC emissions, two scenarios extending until the year 2010 are elaborated in each application sector. Each of these scenarios is realistic.

Scenario I is a business-as-usual scenario and extrapolates present trends. Scenario II is a reduction scenario, in which existing technology potentials for abating or substituting emissions are exploited in each individual HFC sub-sector, without any state regulation of HFC usage. In Scenario II, only emission reduction measures are included whose ecological effect is not neutralized by disadvantages elsewhere.

In Aggregate Scenario I - composed of six sectoral business-as-usual scenarios emissions rise to more than 18,800 t by 2010. In Aggregate Scenario II, emissions rise over the same period to only just under 10,000 t (53%). In terms of global warming impact, Scenario I leads to 19.8 million t CO_2 equivalent, Scenario II only to 10.8 million t. This means a reduction by 9 million t CO_2 equivalent or 45%.

1 2010 Business-as-Usual Emissions Forecast - Scenario I

Scenario I is based for 1995-2010 upon the following sector-specific assumptions:³

- 1. In <u>refrigeration and stationary air conditioning</u> the entire HCFC stock is replaced by HFCs. Substitution is completed by the year 2010, with a refrigerant stock in that year of 30,000 t. The present annual emission rates and recovery ratios in disposal remain constant until 2010.
- 2. In <u>mobile air conditioning</u> the growth in car air-conditioning (A/C) continues until the entire vehicle fleet has reached an A/C ratio of 75%. At this ratio, the refrigerant stock amounts to just under 30,000 t HFC (134a). Charges of air-conditioning units drop annually by 1%, while emission and recovery rates remain at present levels until 2010.
- 3. In <u>PU one component foam</u> the annual growth of can sales in Germany figures 5%. The propellant ratio between 134a and 152a is 2 to 1. The average HFC can content drops continuously from 75 to 66 grams.
- 4. In products of PU foam the blowing agent HCFC-141b is replaced in small-scale applications from the year 2004 onwards at a ratio of 1 to 1 by new liquid HFCs. In PU sandwich panels, HCFCs are replaced in 50% of cases by CO₂ with HFC co-blowing agents. (The other 50% do not use halogenated blowing agents.)
- 5. In <u>XPS rigid foam</u> insulation panels, all manufacturers in Germany convert from the year 2000 onwards from HCFC blowing agents to the HFCs 134a and 152a.
- 6. In propellants for <u>metered dose inhalers</u> CFCs are replaced 100% by the year 2004 by the HFCs 134a and 227. Propellant quantities remain equal.
- 7. In <u>semiconductor manufacturing</u> HFC-23 emissions rise to the same degree as the 6% annual growth of consumption in Germany for dry etching.

³ The sub-sector 'other HFCs' is not examined in two scenarios. Its emissions are equal in both aggregate scenarios.

Table 1: 1995-2010 sectoral HFC emissions								
in business-as-usual Scenario I [t HFC per annum]								
1995 2000 2005 2010								
1. Refrig. and stationary A/C	126	1219	2457	3180				
2. Mobile A/C	126	1016	2352	3611				
3. One component PU foam	1680	1958	2437	3000				
4. Products of PU foam	0	197	495	679				
5. XPS rigid foam	0	4654	6013	7745				
6. Metered dose inhalers	0	120	400	400				
7. Semiconductor manufacturing	3	5	6	8				
8. Other HFC applications	200	129	201	202				
Total	2135	9298	14361	18825				

Under these assumptions, the emissions shown in Table 1 result for the 1995-2010 period. In the calculation, they grow over that period from 2135 to 18,825 tonnes - almost 9-fold.

- By far the most HFC emissions, namely 7745 t or 41%, are emitted in the year 2010 from XPS foam production facilities (Table 1, line 5). 20% of this is HFC-134a, 80% HFC-152a. The ratio between the emissions of these two HFCs differs greatly from their ratio in consumption, because HFC-152a is emitted completely in the first year of its use, while only 25% of HFC-134a is released in the first year, the greater part remaining in the product.
- Roughly 3000 t HFCs are emitted in the year 2010 from each of the three sectors refrigeration and stationary air conditioning (3180 t), mobile air conditioning (3611 t) and one component PU foam (3000 t). These three sectors account for 17%, 19% and 16% of all HFC emissions. Mobile A/C systems contain exclusively HFC-134a. In one component PU foam HFC-134a this makes up two thirds, the rest being HFC-152a. Among the major HFC applications, solely 'refrigeration and stationary A/C' is not a domain of 134a or 152a. Here blends of defined composition predominate, such as 404A/507, but also 410A and 407C. Their GWP is above average.
- Table 2 (last column) shows that in 2010 the CO₂ equivalent emissions from refrigeration and stationary A/C are, at 7.66 million t, more than twice as high as those from XPS foams at 3.04 million t. When expressed in HFC emissions, the ratio is the opposite. Then XPS foams emit 7745 t and refrigeration and stationary A/C only half as much: 3180 t. This is because in XPS foams HFC-152a is used in large quantities, which has a relatively low GWP of 140. In refrigeration systems, the blend 404A/507 predominates, with a 23-fold higher GWP of 3268.
- PU foam products (integral skin foam, rigid foam in small-scale applications and in sandwich panels) and metered dose inhalers are, with emissions of 679 t and 400 t (2010), smaller HFC sectors. Nonetheless, their global warming contribution each amounts to 0.68 million t CO₂ equivalent together more than 1.3 million t. PU foam contains in addition to HFC-134a the newly developed HFC-245fa/365mfc (GWP 815), metered dose inhalers contain HFC-134a or HFC-227 (GWP 2900).

Table 2: 1995-2010 sectoral HFC emissions								
in business-as-usual Scenario I [million t CO ₂ equivalent per annum]								
1995 2000 2005 2010								
1. Refrig. and stationary A/C	0.28	2.90	5.93	7.66				
2. Mobile A/C	0.16	1.32	3.06	4.69				
3. One component PU foam	1.36	1.74	2.17	2.74				
4. Products of PU foam	0	0.26	0.51	0.68				
5. XPS rigid foam	0	1.73	2.31	3.04				
6. Metered dose inhalers	0	0.19	0.68	0.68				
7. Semiconductor manufacturing	0.04	0.05	0.07	0.10				
8. Other HFC applications	1.29*	0.16	0.25	0.25				
Total	2080	8.35	14.98	19.84				

* The high value is due to release of 100 t HFC-23 with the high GWP of 11,700 as a byproduct of HCFC-22 synthesis. These releases last arose in 1995 (until the middle of the year).

'Other HFCs' include HFC emissions as propellants and solvents in industrial aerosols, for fire protection and (only until mid-1995) as a by-product of HCFC-22 synthesis. The global warming impact was very large in 1995 due to this by-product (cf. Table 2). In 2010, the impact is much smaller, at 0.25 million t CO₂ equivalent. The only sector with smaller emissions is that of HFC-23 from semiconductor manufacturing, at 0.10 million t CO₂ equivalent. However - and this is a fundamental reservation - it cannot be excluded that by the year 2010 further HFC applications emerge that are not yet known today.

2 2010 Relative Emission Reductions - Scenario II

The reduction scenario for 1995-2010 is based on the following sectoral assumptions:

- 1. In <u>refrigeration and stationary A/C</u> regular maintenance is mandated by statute from the year 2003 onwards for equipment with charges > 1 kg refrigerant. This cuts both operating emissions and disposal emissions from the serviced equipment by half.
- 2. In <u>mobile A/C</u> (road vehicles) HFC systems are generally replaced by systems using carbon dioxide as refrigerant over the 2007-2009 period.
- 3. In <u>one component PU foam</u> a gradual transition takes place to halogen-free (flammable) hydrocarbon propellants, down to a remainder of 5% HFCs for safety-relevant applications.
- 4. In products of PU foam the HCFC-141b blowing agent is only replaced by half by liquid HFCs in small-scale applications by 2004. In sandwich panels, CO₂ blowing agents with HFC co-blowing agent are only introduced in 25% of cases (instead of 50%). The alternatives in both applications are pentane or CO₂ without HFC.
- 5. In <u>XPS rigid foam</u> insulation panels, almost half of production in Germany already uses from 2000 onwards CO₂ plus ethanol as blowing agent instead of HFCs.
- 6. In <u>metered dose inhalers</u> only 25% of CFC usage is replaced by HFCs. Dry powder inhalers establish themselves as the main form of medication.
- 7. In <u>semiconductor manufacturing</u> new etching chambers are installed exclusively with high-efficiency exhaust purification from 2000 onwards. From 2009 onwards, old equipment is replaced by new systems with exhaust HFC destruction systems.

Under these conditions, the emissions shown in Table 3 result for all sectors together for the period until 2010. Over the 1995-2010 period, emissions only rise from 2135 t to 9984 t. This is about 47% less than in Scenario I in the year 2010.

Table 3: 1995-2010 sectoral HFC emissions								
in reduction Scenario II [t HFC per annum]								
1995 2000 2005 2010								
1. Refrig. and stationary A/C	126	1219	1800	1721				
2. Mobile A/C	126	1016	2322	2969				
3. One component PU foam	1680	1958	1053	150				
4. Products of PU foam	0	197	413	426				
5. XPS rigid foam	0	2653	3427	4415				
6. Metered dose inhalers	0	120	200	100				
7. Semiconductor manufacturing	3	4	4	1				
8. Other HFC applications	200	129	201	202				
Total	2135	7294	9421	9984				

- In Scenario II, too, XPS foam is the largest source of HFC emissions (4415 t) in 2010, although emissions are 43% lower than in Scenario I.
- Mobile air conditioning is the second largest emitter (2969 t). Its relatively small emissions reduction, by 18% compared to Scenario I in the year 2010, is due to the late introduction of HFC-free systems, commencing in 2007. Ten years later, emissions from mobile A/C could already be the smallest of all HFC emission sectors - tending towards zero.

Table 4: 1995-2010 sectoral HFC emissions								
in reduction Scenario II [million t CO ₂ equivalent per annum]								
1995 2000 2005 2010								
1. Refrig. and stationary A/C	0.28	2.90	4.30	4.11				
2. Mobile A/C	0.16	1.32	3.02	3.86				
3. One component PU foam	1.36	1.74	0.96	0.19				
4. Products of PU foam	0	0.26	0.43	0.45				
5. XPS rigid foam	0	0.99	1.32	1.73				
6. Metered dose inhalers	0	0.19	0.34	0.17				
7. Semiconductor manufacturing	0.04	0.05	0.05	0.01				
8. Other HFC applications	1.29	0.16	0.25	0.25				
Total	3.13	7.59	10.67	10.77				

- In terms of global warming contribution (cf. Table 4) 'refrigeration and stationary air conditioning' remains the largest emitter in the year 2010, although emissions drop from 7.66 million t CO₂ equivalent (Scenario I) to 4.11 million t.
- Emissions from one component PU foam experience the greatest reduction in Scenario II, by more than 90%. Instead of 3000 t HFC, only 150 t are used.
- Compared to emissions in Scenario I, emissions from metered dose inhalers are cut down to 25%, from semiconductor manufacturing even to 10%. In products of PU foam, the drop in emissions is less marked.

The different aggregate evolution of emissions in Scenarios I and II over the entire 1995-2010 period is visualized in Diagrams 1 and 2:



Diagram 1: 1995-2010 HFC emissions in Germany in Scenarios I and II in thousands of tonnes. After the sudden rise around the year 2000 due to HCFC replacement, emissions develop in Scenario II at a relatively constant level. This establishes by the year 2010 a growing difference to emissions in Scenario I (reduction potential).

Diagram 2 shows a rise in CO_2 equivalent emissions in Scenario I to almost 20 million t. In reduction Scenario II, emissions in 2010 are only slightly in excess of 10 million t.



Diagram 2: 1995-2010 HFC emissions in Scenarios I and II in million t CO_2 equivalent. In Scenario I, emissions rise by 2010 to almost 20 million t CO_2 equivalent. In Scenario II, they only rise to 54% of this in the year 2010.

Annex: The years 1995 to 1997

Tables 5 and 6 serve to complete the environmentally relevant emission data for the years 1996 and 1997.

1995-1997 emissions in t per annum

Table 5: 1995-1997 sectoral HFC emissions in t/a						
	1995	1996	1997			
1. Refrig. and stationary A/C	126	289	510			
2. Mobile A/C	126	233	386			
3. One component PU foam	1680	1749	1805			
4. PU integral skin foam	0	60	70			
5. Metered dose inhalers	0	0.2	9			
6. Semiconductor manufacturing	3	4	4			
7. Other HFC applications	200	100	100			
Total	4130	4433	4881			

1995-1997 global warming contribution

Table 6: 1995-1997 sectoral HFC emissions in million t CO ₂ equivalent							
1995 1996 1997							
1. Refrig. and stationary A/C	0.28	0.65	1.18				
2. Mobile A/C	0.16	0.30	0.50				
3. One component PU foam	1.36	1.39	1.48				
4. PU integral skin foam	0	0.08	0.09				
5. Metered dose inhalers	0	0.00	0.01				
6. Semiconductor manufacturing	0.04	0.05	0.05				
7. Other HFC applications	1.29	0.12	0.12				
Total	3.13	2.58	3.45				

1 Refrigeration and Stationary Air-Conditioning

This section is concerned, firstly, with stationary and mobile refrigeration equipment for goods and processes and, secondly, with stationary air-conditioning systems for buildings (mobile air-conditioning systems are treated in the 2nd section). This field can be disaggregated into eight sectors, listed in the following in the estimated order of their overall stock of halogenated refrigerants (CFCs, HCFCs, HFCs) in Germany:

- 1. Commercial refrigeration (approx. 40%).
- 2. Middle industrial refrigeration (reciprocating compressors) (20%).
- 3. Small commercial and industrial hermetics < 1 kg refrigerant (20%).
- 4. Large industrial refrigeration and air-conditioning (6-7%).
- 5. Room air-conditioners < 2 kg refrigerant (5%).
- 6. Middle air-conditioning (reciprocating compressors) (3-4%).
- 7. Transport refrigeration (2-3%).
- 8. Domestic heat pumps < 2 kg refrigerant (ca. 1%).
- Before 1991, three refrigerants were essentially used in the eight sectors. CFC-12 (R-12) was used in the normal temperature range (0°C/+10°C), and the CFC/HCFC blend R-502 at low temperatures (-25°C/-10°C). The quantitatively most important refrigerant, HCFC-22 (R-22), was used at medium temperatures and also in the majority of air-conditioning systems.
- Due to their similar chemical-physical properties, HFCs are the substances of choice to substitute the above chlorine-containing ozone-depleting substances. However, halogen-free substances also come into question as substitutes. In the course of CFC phase-out, substances and processes which had been displaced and neglected for decades due to the favourable inherent properties of CFCs (nonflammability, nonpolarity, nontoxicity) were rediscovered and further developed throughout the second half of the 1980s. This led to a switch from numerous former CFC applications to processes and substances with neither ozone-depleting nor global warming impacts.
- As in other sectors, too, a search is under way in the refrigeration sector for alternatives that promise long-term security of supply. Here HFCs are exposed in some areas to competition from the refrigerants ammonia, hydrocarbons and CO₂. The above list of eight refrigeration and air-conditioning sub-sectors thus lacks one which accounts for up to 20% of the refrigerant stock in other industrialized countries: domestic refrigeration. In Germany, the refrigerant used here is isobutane (R600a), R-134a having only played an interim role until 1994/95.⁴
- In the other refrigeration and air-conditioning sectors, no comparable abandonment of an entire area of HFC application has taken place, nor is this currently to be expected. As a rule of thumb we can say: R-12 was and continues to be substituted by HFC-134a, and R-502 and R-22 were and continue to be substituted by the HFC blends R-404A/R-507. The HFC blends 407C or 410A are suited for some R-22

⁴ The HFC refrigerant stock in Germany additionally contains these household appliances, with a bank of approx. 400 t 134a. In view of their low operating emissions (<1%) they are not treated further in the present study.

refrigeration applications and most R-22 air-conditioning applications. In the eight sectors, the four HFC refrigerants - 134a, 404A/507, 407C and 410A - will substitute the bulk of CFCs and HCFCs over the medium term.

 Beyond the issue of market availability of HFCs, the speed and timing of CFC substitution by HFCs depend upon technological and political factors.

On the <u>technological</u> side, in existing refrigeration and air-conditioning equipment it rarely suffices for CFC substitution by HFCs to simply switch the refrigerant. For HFC operation, numerous components and ancillary process inputs need to be modified, so that the use of HFCs practically or effectively requires new equipment. Pure HFCs are therefore generally only used when an old refrigeration or air-conditioning system is replaced by a new one.

In areas where the legislator required CFC phase-out in still intact systems, such as in Germany for the refrigerant R-12 by the end of 1995, the operators of these systems did not undertake advance investments in HFC-ready equipment to any great extent. They rather took up offers of the chemical industry to continue to operate old equipment until the end of its service life with drop-in refrigerants (Schwarz/Leisewitz 1998). These are refrigerant blends which, while usually containing HFCs, predominantly consist of HCFCs, notably R-22

The <u>political</u> factor is the nationally applicable CFC phase-out schedule. Thus all fully halogenated CFCs (R-12, R-502) are banned since 1995 throughout the European Union (EU), but under new EU rules (EC 98) the partially halogenated CFCs are only banned for new refrigeration equipment from the year 2001 onwards, and for refilling existing equipment from 2010 onwards. In Germany, in contrast, HCFC-22 is already banned for new equipment by the CFC-Halon Ban Ordinance from 1.1.2000 onwards.

• In Germany, where from 1999 onwards none of the three traditional refrigerants can be used any longer in new equipment, the pressure upon refrigeration equipment manufacturers is particularly high to put substitute substances on the market in time, and thus to accelerate the introduction of HFCs.

1 1995-1997 New Consumption and Emissions in Germany

- The annual <u>new consumption</u> of HFC refrigerants for applications within Germany doubled over the 1995-1997 period, from 1144 t to 2418 t (Refrigerant Producer Survey).
- Pure 134a and the blends 404A and 507 the latter both consisting mainly of 125 and 143a and treated in this study as a uniform refrigerant - are by far the most important pure HFC refrigerants. Together, the share of 134a and 404A/507 in annual new consumption figures more than 90%. The remainder is composed to a smaller part of the refrigerants R-23, R-407C and R-410A and to a larger part of such HFCs which are only components in R-22-based drop-in refrigerants (cf. Table 7).
- Of the two main refrigerants, 404A/507 has experienced a stronger growth than 134a. In 1995, consumption of the former within Germany was still significantly

lower than that of the latter, in 1996 it had reached the same level, and since 1997 404A/507 accounts for the largest proportion of refrigerant consumption, at a level of 1141 t (see Table 7).

Table 7: 1995-1997 HFC refrigerant consumption and operating emissionswithin Germany from refrigeration and stationary air-conditioning equipment,								
ex	pressed in	t refrigera	nt and in m	illion t CO	₂ equiv.			
Refrigerant	1995 1996 1997					97		
	Consumption	Emission	Consumption	Emission	Consumption	Emission		
134a	667	65	878	142	1067	229		
404A/507	369	38	877	105	1141	215		
Other HFCs*	108	23	130	42	210	66		
Total in t	1144	126	1885	289	2418	510		
Mill. t CO₂ equiv.		0.28		0.65		1.18		

* 407C, 410A, 23; drop-in components: 125 in 402A (60%), 408A (7%), 152a in 401A/B (13%), 143a in 408A (46%).

• This has the following causes on the side of R-134a and on the side of R-404A/507:

<u>Firstly</u>, since 1996 134a has not been able to assume the expected role as a substitute for CFC-12, which at that time was to be replaced in existing equipment within a period of two years (by the middle of 1998). Other than in large centrifugal compressor systems, approx. 50% of R-12 in commercial refrigeration was replaced by R-22-containing drop-in refrigerants - above all by R-401A, of which only 13% consists of HFCs (152a) (Schwarz/Leisewitz 1998, VI).

<u>Secondly</u>, the impending ban deadline for R-22 (1.1.2000) sensitized purchasers of new equipment replacing R-22 equipment in terms of longer-term product security. In competition with comparable equipment using 404A/507, R-22 equipment has scarcely any market prospects. The strong growth of 404A/507 is to a considerable extent at the cost of R-22.

- Although 407C and 410A are also substitutes for R-22 predominantly in the airconditioning sector - their consumption remains low. This is an outcome of the global market for air-conditioning systems and equipment, which is dominated by large North American and Far East companies. The European market only accounts for 6% of the global air-conditioning equipment market (Bailly 1998), and, in contrast to Germany, R-22 will continue to be permitted for a long time in the major buyer countries. Moreover, 410A requires new, more pressure-resistant components, which have only became available recently.
- If we apply specific emission rates⁵ to the eight refrigerant stocks of the years 1995 to 1997⁶ which result from the accumulated consumption of the years 1993 to 1997 (Refrigerant Producer Survey), then we receive the estimates for operating emissions of HFCs shown in Table 7. In total, these have risen from 126 t (1995) over 289 t (1996) to 510 t in the year 1997 (cf. Diagram 3). Both new consumption and

⁵ On their determination see the next section.

⁶ According to the draft inventory guidelines of the IPCC, the total of half of the stock of the year n-1 and half of the stock of the year n is the reference quantity for emission rates by which to estimate operating emissions of the year n (IPCC 1999).

emissions are comprised almost 90% of R-134a and R-404A/507, the two refrigerants being at roughly equal levels.

- Commercial refrigeration is the largest individual emitter, accounting for approx. 77% of HFC emissions in 1997. This is followed in second place by transport refrigeration with 5% of total emissions. These are both sectors with high emission rates (20% and 15%, respectively).
- The CO₂ equivalent contribution to global warming of all eight sectors rose over the 1995-1997 period from 0.28 to 1.18 million tonnes - see Diagram 3. (The estimate for 1998 is 1.73 million t. CO₂ equiv.)



Diagram 3: HFC emissions from refrigeration and stationary air-conditioning in 1993-1998, shown in tonnes of HFC refrigerant and CO_2 equivalent. HFC emissions (left scale) rose in the 1995-1997 period from 126 to 510 t (columns). The CO_2 equivalent contribution to global warming (right scale) rose accordingly from 0.28 to 1.18 million t (line). The diagram also shows the estimated values for 1998: 746 t HFCs and 1.73 million t CO_2 equivalent.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

Forecasting estimates of HFC emissions from the refrigeration and stationary airconditioning sectors requires the following basic data:

- Size of sectoral HFC stocks after complete CFC and HCFC substitution. Breakdown of these stocks among the various HFC refrigerants.
- Sector-specific operating emission rates.
- Refrigerant recovery rates on disposal.
- Mean equipment operating life (because of the point in time of the first disposal emissions).

Table 8: Basic data for determination of HFC emissions from refrigeration and							
Application	lationaly a	Refrigerant	1997	stock		stock	
1 Domestic heat n	umns (SI	15a)					
$\sim 1.5 \text{ kg}$	FR 3 3%	407C	22 t	80%	250 t	80%	
~ 0.6 kg	ER 2.5%	410A		20%	2001	20%	
2 Commercial refr	ideration	(SI 10a)			J []		
New equipment	FR 15%	134a	2774 t	44%	12450 t	32%	
		4044/507	21140	56%	124001	60%	
		407C		0.2%		4%	
		410A		0%		4%	
Old equipment	FR 20%	152a 401	507 t	11%	0 t	0%	
	211 2070	125402/408	0011	51%		0%	
		143a 408		8%		0%	
		134a 413		30%		0%	
3 Large refrig and	l air-cond	(SI 252)			J []		
Centrifugal compr		13/12	515 t	00%	2000 t	50%	
Screw compressor		104a 407C	5151	1%	2000 (25%	
		4070		0%		25%	
		4107		070		2370	
4. Middle industria	l refrig. (S	SL 15a)			л г 		
Recipr. compr.	ER 6%	134a	418 t	30%	6000 t	16%	
		404A/507		62%		33%	
		23		5%	_	1%	
		407C		3%	_	17%	
		410A		0%		33%	
5. Middle air-condi	tioning (S	L 15a)					
Recipr. compr.	ER 5%	407C	6 t	100%	1000 t	50%	
		410A		0%		50%	
6. Room air-condit	ioners (SI	_ 15a)					
~ 1.5 kg	ER 2%	407C	6 t	100%	1500 t	40%	
		410A		0%		60%	
7 Small ind /comm	borm (S	SI 102)			J []		
	FR 1 5%	13/12	1150 t	19.5%	6000 t	/7%	
		1044/507	11001	49.5%	00001	<u> </u>	
		407C		1%		-170 6%	
		4070		170		070	
8. Transport refrig		404	450.4	000/		000/	
I FUCK COOLING SL10a	ER 20%	134a	150 t	33%	350 t	29%	
		404A/507		59%		54%	
		410A		1%		1/%	
Orntoi		1528		1%	070 /	0%	
Containers SL15a	ER 10%	134a	96 t	100%	3/0 t	100%	
Snip cooling SL30a	ER 25%	410A	0 t	0%	80 t	100%	
Total/average	ER 9%		5644 t		30000 t		

Abbreviations: ER = operating Emission Rate per year; SL = Service Life in years (a). Explanations are given in the following text.

The presentation of figures in Table 8 is a refinement of the scheme used in the 1996 report (Schwarz/Leisewitz 1996, 91).

With the exception of the recovery rate, for which, lacking an empirical basis, a default value of 70% of the original refrigerant charge is generally assumed, as proposed by the IPCC (IPCC 99), Table 8 shows all of the determinants (basic data) on which the year-2010 emissions model is based. The transparency thus provided is intended to make the forecast data reproducible.

Both the disaggregation of the overall stock into <u>eight sub-sectors</u> and the determination of <u>absolute quantities</u> and of internal <u>refrigerant compositions</u> of the eight subsector stocks made use not only of national information (Basic Data Update 1999), but also evaluated numerous recently published international studies (UNEP-RTOC 1998; March 1998, 1999; AFCE 1998; DTI 1998; ECOFYS 1999). The determination of the <u>emission rates</u> also made use of the default values considered by the IPPC (1999) as typical for the various applications.

Nonetheless, the emission rates need to be viewed with due caution. The wide variance of emission rates for identical applications that is to be found in the literature is only partly an outcome of technological changes in equipment leak-tightness or an expression of national differences. It is to a great degree an outcome of real uncertainty, as robust empirical research, based for instance on monitoring refill quantities over a period of several years, has been carried out practically nowhere (cf. Johnson 1999).

The present study cannot remedy this deficiency. However, the emission rates refer to new equipment (second half of the 1990s) and are situated in the lower range of the default values of the IPCC and UNEP-RTOC. They are rather on the conservative side.

• Table 9 shows for individual years and Diagram 4 for the whole 1995-2010 period the HFC emissions that result without revolutionary technological improvement of equipment leak-tightness and without state regulation. This Scenario I is based on the data presented in Table 8, in particular on constancy of the emission and recovery rates.⁷

Table 9: 1995-2010 HFC refrigerant emissions from refrigeration and stationary							
air-conditioning in Business-as-Usual Scenario I							
1995 2000 2005 2010							
Operating emissions	126	1199	2216	2606			
Disposal emissions	0	19	241	574			
Total emissions in t	126	1219	2457	3180			
Ems. in million t CO₂ equivalent	0.28	2.90	5.93	7.66			

• The process of HCFC substitution will take until the year 2010. The eight refrigerant stocks, together comprising 30,000 t, will then consist exclusively of HFCs. Operat-

⁷ In contrast to other emissions forecasts, the emission rates taken here are <u>not</u> reduced by the year 2010, but held constant. This is because the improved leak-tightness of new equipment that is to be expected over the next ten years is cancelled out by the increasing leakage risks caused by the ageing of equipment installed since 1995.

ing emissions will then figure 2606 t, corresponding to an average annual emission rate of 8.7%. Operating emissions (leaks, maintenance losses) will peak in the following year.⁸ These are joined by disposal emissions (recovery losses during scrapping) amounting to 574 t. The two make up the total emissions of 3180 t. If we place these in relation to the refrigerant stock of 30,000 t, then the rate is 10.6%.



Diagram 4: Operating and disposal emissions of HFC refrigerants from refrigeration and stationary air-conditioning in 1995-2010 in t (Scenario I). Operating emissions (lower area) rise until the year 2005 to a level in excess of 2200 t. Their growth then slows until the year 2010. This is because, firstly, applications with high emissions such as commercial refrigeration have already reached their peak and, secondly, the stock grows mainly in the form of air-conditioning equipment charged with 407C- and 410A, which have lower emission rates. Disposal emissions (upper area) only grow substantially after 2003 when the first units are retired.

- While operating emissions remain stable in the period after 2010, disposal emissions continue to rise for approximately a further ten years. This results firstly from the differing operating lives of HFC-filled equipment and, secondly, from the differing times of their introduction. New medium-sized and room air-conditioning systems only contain the HFC blends 407C or 410A instead of HCFC-22 since the end of the 1990s. These systems will not yet give rise to disposal emissions by 2010. In contrast, disposal emissions already set in from 2003 onwards from commercial and transport refrigeration, from small and commercial and industrial hermetics and from centrifugal compressors retrofitted since 1995 from R-12 to R-134a. These systems were not only charged earlier with HFCs, but also have a short (remaining) operating life.
- Disposal emissions <u>prior to</u> 2003 (Table 9 lists these as amounting to 19 t in 2000) are of a special kind. These are recovery losses arising during the scrapping of

⁸ Emission rates refer to half of the stock of the previous year and half of the stock of the current year.

retired equipment which was filled in the years 1994 to 1999 not with pure HFCs but with HFC-containing drop-in refrigerants in the course of CFC-12 substitution. Emissions of the HFCs 125, 152a and 143a serving as drop-in blend components arise from 1994 onwards during operations and from 1999 onwards also during scrapping. From 2005 onwards, they no longer play any role.

• The global warming contribution, expressed in CO₂ equivalent, rises from 0.28 million tonnes in 1995 to 7.66 million tonnes in 2010 (cf. Table 9). (From this we may calculate an average GWP of the refrigerants emitted of just under 2400.)

3 2010 Relative Emission Reductions - Scenario II

- The debate on refrigerant substitutes with a low global warming potential currently concentrates on hydrocarbons (HCs), ammonia and CO₂. Ammonia (NH₃) will doubtlessly expand its leading role in industrial refrigeration, but this growth is already taken into consideration in Scenario I. A breakthrough of CO₂ technology before 2010 can be expected most in car air-conditioning, but experiments are also under way in heat pumps and transport containers (Süß 1999). As concerns HCs, these have proven themselves in small hermetic compressors domestic appliances, heat pumps and small room air-conditioners.
- It is presently not to be expected that HCs will gain widespread application in the large sectors of commercial and industrial refrigeration and building air-conditioning by 2010. After initial tests carried out by a number of food retail chains with primary HC refrigerant loop and indirect cooling, the euphoria over flammable refrigerants for larger systems has sobered down. As yet, the higher energy requirement of indirect systems is not only more expensive, but contributes - at least in Germany overall through more CO₂ more (> 10%) to global warming than direct systems (TEWI perspective). This is the experience of operators of indirectly cooling HC supermarket systems (Kalter/Boergen 1998).
- Given a lack of alternative refrigerants with which to substitute HFCs, the reduction
 of emissions is all the more important. Many countries have adopted regulations
 governing refrigerant recovery (UNEP-RTOC 1998, 242). Most, however, refer
 exclusively to chlorine-containing substances (ODS). This also applies to the
 proposal of the European Commission (EC 98), which provides for annual
 monitoring of equipment tightness for ODS.
- In the Netherlands, practical experience has been made with the influence of statutory stipulations upon refrigerant emissions from stationary refrigeration. There the government adopted in 1994 regulations governing leak-tightness of refrigeration equipment which cover explicitly all halogenated refrigerants, not only ODS (Technical requirements for refrigeration equipment / Regeling lekdichtheidsvoorschriften koelinstallaties 1994). These requirements, modelled on a draft European Standard (prEN 378), which concern above all units > 3 kg refrigerant charge (but also smaller units if they have a refrigeration capacity > 500 watt), essentially have two parts: Firstly, technical rules for construction and installation (materials, components, types of connections etc.) and, secondly, rules on regular

inspection and maintenance by operators and external technicians. Both parts are supervised by STEK, an independent foundation (Technical Requirements 1994).

- First evaluations show that, since 1994, refrigerant emissions have dropped distinctly in the Netherlands for equipment that falls under the new rules. For instance, the annual losses of 75 supermarkets have fallen from 15% to 3% (van Gerwen et al. 1998). Both van Gerwen (1999) and Graal (1999) attribute the emissions reduction half to new technological requirements and half to regular maintenance by external technicians.
- In Germany, the federation of refrigeration equipment crafts (Verband des Kälteanlagenbauerhandwerks, VDKF) has been calling for mandatory maintenance of refrigeration and air-conditioning equipment since 1996 (VDKF 1996). At a meeting on "Measures for the regular maintenance of refrigeration and airconditioning equipment" held at the German Federal Environmental Agency (Umweltbundesamt, UBA) in Berlin on 13.3.1996, the crafts representatives pointed out the savings in refrigerants and energy consumption that regular maintenance can provide, thus covering the additional costs. The operator side, however, viewed cost-neutrality sceptically.
- It would be possible to mandate by statute the regular maintenance of refrigeration equipment in Germany on the basis of the Chemicals Act (Chemikaliengesetz), or on the basis of the Emission Control Act (Immissionsschutzgesetz). <u>This would not</u> <u>be a measure aimed at preventing the use of HFCs, but would rather presuppose</u> <u>such use</u>.
- Within only a few years, such a measure could cut emissions from serviced refrigeration equipment, if not down to 25%, at least to half. It needs to be kept in mind that the mandatory maintenance called for the by the refrigeration crafts is only one of the two elements of the Dutch rules, which further stipulate technological specifications for component design, installation etc.
- For the relative emission reductions (Scenario II) it is assumed that from 2003 onwards mandatory maintenance applies in Germany to all refrigeration and air-conditioning equipment.
- The only exemptions to this requirement would be modelled on the well-proven de minimis threshold for R-12 substitution in old equipment - stand-alone hermetics in which the refrigerant is cycled in a permanently closed loop, and all equipment with charges smaller than 1 kg. The requirement thus applies to 75% of the total refrigerant stock in refrigeration and air-conditioning equipment. Only the following would be exempted from mandatory maintenance: small commercial and industrial refrigeration (6000 t stock in 2010), most room air conditioners (1500 t stock in 2010) and most domestic heat pumps (250 t stock in 2010).
- The assumed effect is that both operating emissions and recovery losses drop gradually for serviced equipment, both being 50% lower than in Scenario I within five years at the latest. The values shown in Table 10 and Diagram 5 result:

Table 10: 1995-2010 HFC refrigerant emissions from refrigeration and								
stationary air-conditioning in Scenario II								
1995 2000 2005 2010								
Operating emissions	126	1199	1579	1367				
Disposal emissions	0	19	221	355				
Total emissions in t	126	1219	1800	1721				
Ems. in million t CO₂ equivalent	0.28	2.90	4.30	4.11				

- After introduction of mandatory maintenance (2003), in the year 2010 operating emissions only figure 1367 t (52%) compared to 2606 t in Scenario I, and disposal losses only 355 t (62%) instead of 574 t. Total emissions are not 3180, but only 1721 t (54%). The emission rate (total emissions) over the total refrigerant stock of 30 000 t figures 5.7%. This is not down by half compared to the 10.6% rate in Scenario I, as 25% of the refrigerant stock is exempted from mandatory maintenance, but is less by 45%.
- Diagram 5 shows the emissions curve in Scenario II. The outcome of mandatory maintenance is that from the time of its introduction onwards, emissions (total emissions) stop rising as in Scenario I, remaining instead at a roughly constant level.



Diagram 5: Operating emissions and disposal emissions of HFCs from refrigeration and stationary air-conditioning in 1995-2010 in t - Scenarios I and II. Instead of rising as in Scenario I (being all the areas under the uppermost curve), emissions level out after 2003. The sum of operating and disposal emissions (shaded areas) remains relatively constant until 2010 at a much lower level.

• The global warming contribution drops by 3.55 million t CO₂ equivalent: from 7.66 million to 4.11 million t. Diagram 6 shows the curves for Scenarios I and II.



Diagram 6: HFC emissions from refrigeration and stationary air-conditioning in million t CO_2 equivalent (Scenarios I and II). From 2003 onwards, the global warming contribution no longer grows in Scenario II - in contrast to Scenario I.

2 Mobile Air-Conditioning

Mobile air-conditioning (MAC) comprises equipment installed in passenger cars, trucks, buses, railcars and on ships. Refrigerant inventories, consumption and emissions are all dominated by cars, with a share of 95% of each. The business-as-usual forecast indicates that by the year 2010, MAC systems in Germany will total an inventory of 29,700 t HFC, which is of the same order as the total of refrigeration and stationary air-conditioning equipment (30,000 t HFC). Of this, cars will account for more than 28,000 t. 'Car air-conditioning (A/C)' and 'other MAC' systems are treated separately in the following.

HFC refrigerants used in MAC systems are almost exclusively R-134a. There are two minor exceptions. The first is that when old R-12 air-conditioning units were retrofitted, a small percentage was filled with the drop-in refrigerant 413A instead of R-134a; how-ever, 413A consists to 88% of R-134a. The second exception is new ship air-condition-ing systems, of which in Germany only half are charged with R-134a, the other half with R-404A/507. However, new usage of R-404A/507 only amounts to 1.5 t annually.

1 1995-1997 New Consumption and Emissions in Germany

In new vehicles charged by the manufacturer and destined for the German market, the process of switching from R-12 to R-134a extended from April 1992 to October 1993. This concerns cars manufactured both in Germany and abroad, and also trucks, buses and railcars. Only the small segment of ship air-conditioning developed differently; here new systems were charged exclusively with R-22 until 1996, and only since 1997 has conversion to R-134a or R-404A got under way (Haug 1999). Aftermarket car systems had switched to R-134a by the end of 1994. In Germany, old vehicles fitted with R-12 air-conditioning units are permitted to continue using these until the end of their service life. Nonetheless, for these old systems, retrofit rates to R-134a figured in the 1995-1998 period some 10% in cars and trucks, 40% in buses and 100% in railcars (Schwarz/Leisewitz 1998). The old systems will retire from the fleet between 2002 and 2005.

MAC systems for road vehicles are not hermetically sealed. Their compressors are driven by the engine and exposed to its vibrations, and their refrigerant flows through flexible piping (hoses). However, emission rates of these systems have dropped in the course of CFC phase-out (Nonnenmann 1995). More impermeable hoses and improved sealing have cut operating losses by half compared to CFC systems. Moreover, in repair and maintenance the refrigerant is no longer discharged entirely to the atmosphere, but is rather suctioned off with a loss of approx. 10% and refilled.

Annual system refrigerant emissions have thus dropped from more than 30% (prior to 1992) to approx. 10% (new systems). Over a system life of 12 years, 10% loss per year corresponds to 120% refrigerant loss, 100% being one system charge. This 120% figure is made up of two half-refills to top up emission losses, and two times 10% maintenance loss⁹. Suction extraction of refrigerant in scrapping, which has been used

⁹ As in all segments of the refrigeration & air-conditioning sector, only expert estimates of emissions are available for MAC systems in cars, but no independent long-term measurements. The best estimates

by repair shops in the past for CFCs, is already equipped to deal with R-134a. This is restricted in Germany by the circumstance that numerous second-hand vehicles are exported to countries to the east, where as yet little attention is given to the refrigerant in scrapping. However, extraction rates can be improved in highly industrialized countries, too. A look at Japan, an island state, is revealing. Here the car industry aims at a recovery rate of 70% from old vehicles with R-134a systems by the year 2010 (Tujibayashi 1999).

Note on emissions:

For mobile air-conditioning systems (cars, trucks, buses, railcars, ships), this study generally assumes annual operating emissions of 10% and a one-off recovery loss upon scrapping (disposal emissions) of 30% of the charge. The operating emissions of a year n are calculated as 10% x (0.5 x inventory_{n-1} + 0.5 x inventory_n).

1.1 Car Air-Conditioning

- The annual new consumption of R-134a for first fill of car air-conditioning (A/C) systems destined for the German market (excl. filling in Germany of cars for export) rose greatly in the 1995-1997 period: from 781 to 1862 t (cf. Table 11).
- This rise was caused by the upswing in A/C penetration (A/C ratio) of annual new car registrations. In 1992, the last full year with CFC refrigerants, the A/C ratio had only figured 9.4% (Schwarz/Leisewitz, loc. cit., 38). Since then, it has risen steeply, first in German car makes, then also in the imported makes. For both together, the A/C ratio had reached 24% by 1995. In 1997, at 52%, it exceeded half of all new car registrations for the first time¹⁰. The rise is continuing. The preliminary A/C ratio for 1998 is 68% (Car Air-Conditioning Survey 1999).

(134a) in car air-conditioning systems in Germany, in t HFC and million t CO ₂								
equivalent								
1995 1996 1997						97		
Refrigerant	Consump.*	Emission	Consump.	Emission	Consump.	Emission		
R-134a	781	113	1349	211	1862	356		
Mill. t CO2 equiv.		0.15		0.27		0.46		
A/C ratio**	24	%	39	%	52	%		

Table 11: 1995-1997 consumption and operating emissions of HFC refrigerants(134a) in car air-conditioning systems in Germany, in t HFC and million t CO2

Source: Car Air-Conditioning Survey 1999.

* Consumption in Germany = First fill of new car air-conditioning systems for the German market incl. imports, excl. exports, excl. top-up to compensate for continuous losses, plus substitution of old R-12 systems within Germany.

** Proportion of air-conditioned vehicles in all new car registrations of a year in Germany.

• The rise in consumption is scarcely reduced by smaller unit charges. In 1995, charges averaged 0.88 kg for German car makes and 0.77 kg for foreign makes. By

include, in our opinion: AFCE 1998; IPCC 1999; UNEP-RTOC 1998; and the contributions by Baker and Preiseggger in: Joint IPCC/TEAP 1999.

¹⁰ In the past, real growth has always exceeded predicted rates: Fischer 1997, Sieverding 1998. Even the 75% A/C ratio in 2002 predicted by Holdack-Janssen 1998 will presumably be reached two years earlier.

1997, these values had dropped to 0.86 and 0.75 kg, respectively. One explanation for this is that more and more smaller cars are being equipped with air-conditioning.

- In the 1995-1997 period, operating emissions rose from 113 to 356 t (cf. Table 11 and Diagram 7, which also shows the preliminary value of 541 t for 1998). Disposal losses did not yet arise. These will only start to arise after 2002, when the old R-12 units retrofitted from 1995 onwards start to be scrapped. The first annual cohorts containing R-134a as first fill will be scrapped in 2005.
- The global warming contribution by emissions rose in the 1995-1997 period from 0.15 to 0.46 million t CO₂ equivalent (Table 11 and Diagram 7).



Diagram 7: 1993-1998 HFC-134a emissions from car air-conditioning systems, in t HFC and million t CO_2 equivalent. The rise in emissions has been unremittingly steep since 1993, both in tonnes HFC-134a (left scale, black columns) and in terms of CO_2 equivalent (right scale, line). The preliminary figure for CO_2 equivalent in 1998 is 0.7 million t.

1.2 Other Mobile Air-Conditioning

- The average charge of truck air-conditioning systems figures 1.0 kg in vehicles below 16 t laden weight, and 1.2 kg in vehicles above 16 t laden weight. New bus air-conditioning systems receive on average a refrigerant charge of 10 kg, railcar systems 18 kg. Ship systems are the largest, with 100 kg charge. As the A/C ratio of these vehicles is rising continuously, annual new consumption of R-134a for first fill of systems in Germany rose over the 1995-1997 period from 59 to 78 t. This was joined by some 20 t annually for retrofitting old R-12 systems (cf. Table 12, line 2).
- Over the 1995-1997 period, operating emissions rose from 13 to 31 t (cf. Table 12, line 2). HFC disposal losses do not yet arise until 2002 apart from accidents and suchlike.
| Table 12: 1995-1997 consumption and operating emissions of HFC refrigerantsin car and other mobile air-conditioning systems in Germany, | | | | | | |
|---|--|------------|--------------------------|-----------|------|------|
| | in t HF | C and mill | lion t CO ₂ e | quivalent | | |
| 1995 1996 1997 | | | | | | |
| | Consump.* Emission Consump. Emission Consump. Emis | | | | | |
| 1. Car A/C | 781 | 113 | 1349 | 211 | 1862 | 356 |
| 2. Other MAC | 72 | 13 | 86 | 21 | 98 | 31 |
| Total | 853 | 126 | 1435 | 233 | 1960 | 386 |
| Mill. t CO ₂ equiv. | | 0.16 | | 0.30 | | 0.50 |

Source: Car Air-Conditioning Survey and Other MAC Survey 1999.

* Consumption in Germany = First fill of new air-conditioning systems for the German market incl. imports, excl. exports, excl. top-up to compensate for continuous losses, plus substitution of old R-12 systems within Germany.

• Over the 1995-1997 period, the global warming contribution of 'other MAC' emissions rose from 0.02 to 0.04 million t CO₂ equivalent (not listed separately in Table 12, but combined with car A/C emissions to give a joint figure).

2 2010 Business-as-Usual Emissions Forecast - Scenario I

In contrast to all other former CFC applications, in the MAC sector the former CFC stock can not be taken as an orientation for forecasting the HFC stock in the year 2010. It is too small for that. In car A/C systems, the R-134a stock had already exceeded by early 1996 the historic peak of R-12 in 1993 (2000 t). Even in terms of global warming impact, the contribution of R-12 at that time (Schwarz/Leisewitz 1995, 23), namely 4.25 million t CO_2 equivalent, will be exceeded by R-134a emissions by the year 2010, even though the GWP of R-134a (1300) is far lower than that of R-12 (8500).

2.1 Car Air-Conditioning

Forecasting the estimated R-134a emissions from car A/C up to the year 2010 (Business-as-Usual - Scenario I) requires a model with <u>dynamic</u> parameters:

- 1. Annual new car registrations average 3.55 million and increase the car fleet from 40.7 million (1995) to 48.6 million (2010) following Shell 1995.
- 2. Two thirds of new registrations are manufactured in Germany, one third abroad.
- 3. The A/C ratio of new registrations reaches 90% in German makes in the year 2001, and then remains constant. In foreign makes, the corresponding plateau is at 80% and applies from 2005 onwards.
- 4. A/C saturation figures 75% of the total fleet in Germany.
- 5. The average charge of new A/C units drops as in the past (Car Air-Conditioning Survey) by 1% annually: from 0.88 kg (1995) to 0.76 kg (2010) in German makes and from 0.75 to 0.66 kg in foreign makes. The overall average charge of all new A/C units drops over that period from 0.86 to 0.73 kg.
- 6. The service life of a new A/C unit figures 12 years.
- 7. The operating emission rate is steady at 10% of the refrigerant stock, according to the equation "10% x (0.5 x stock_{n-1} + 0.5 x stock_n)".

 The rate of refrigerant recovery losses in end-of-life car scrapping is steady at 30%. (This low rate appears possible because annual cohorts of new R-134a units will only be due for disposal from 2005 onwards.)

Table 13 shows for four selected years and Diagram 8 for the entire 1995-2010 period the HFC emissions which follow from the above eight model assumptions. In this 'Scenario I', the refrigerant stock peaks in 2010 at 28,078 t. Operating emissions, which, due to the mode of calculation, peak in the subsequent year, figure 2792 t in 2010. Disposal emissions figure 626 t. The two forms of emission taken together make up the 3418 t total emissions of the year 2010. Based on the refrigerant stock in car A/C systems in that year (28,078 t), the total emission rate is 12.2%.

Table 13: 1995-2010 HFC-134a emissions from car air-conditioning in t HFC-134a and million t CO₂ equivalent in Scenario I						
1995 2000 2005 2010						
Operating emissions [t]	113	955	2150	2792		
Disposal emissions [t]	0	0	74	626		
Total emissions [t] 113 955 2223 3418						
Ems. in mill. t CO ₂ equivalent 0.15 1.24 2.89 4.44						
Stock [t]	1496	10668	22626	28078		

Basis: Car Air-Conditioning Survey 1999. Extrapolation to 2010 based on model assumptions explained in the text.

Outlook after 2010: Because from 2010 onwards the car fleet - according to Shell forecast - will no longer grow significantly and the A/C ratio of the fleet - according to the model assumptions made here - will level out at 75%, operating emissions will drop slightly from 2011 onwards.



Diagram 8: 1995-2010 R-134a operating and disposal emissions from car air-conditioning in t R-134a (Scenario I - Business-as-Usual). Operating emissions (lower area) reach 2792 t in 2010 (peak in 2011). Disposal emissions (upper area) commence in 2003 (retirement of retrofitted old R-12 units) and become substantial from 2006 onwards, reaching 626 t in 2010. Calculated total emissions are then 3418 t.

The drop in operating emissions after 2011 is due to the continuous reduction of unit charges by 1% annually. Disposal emissions, in contrast, continue to rise slightly - peaking in 2016 at 760 t. Total emissions peak in 2013 at 3532 t.

Over the 1995-2010 period, the global warming contribution rises from 0.15 to 4.44 million t CO_2 equivalent (cf. Table 13). Owing to the lower GWP of HFC-134a, this is substantially less than in the refrigeration & stationary air-conditioning sector.

2.2 Other Mobile Air-Conditioning

The emission forecast for 'other MAC' (trucks, buses, railcars and ships) is based on the parameters shown in Table 14, which are not commented upon further here.

Table 14: Parameters for the development of 'other MAC' systems						
(i.e. excluding cars) in 1995-2010						
	New vehicles	A/C	ratio	Charge	Target stock	
	reg. annually	in Ge	rmany	per A/C	in 2010	
	in Germany			unit		
		1995	2010			
Trucks (SL 10a)						
< 2 t laden weight	150000	8%	38%	1.0 kg	435 t	
2-16 t	50000	5%	35%	1.0 kg	130 t	
> 16 t	18000	36%	90%	1.2 kg	370 t	
Buses and coache	s (SL 15a)					
Urban	2400	14%	63%	10 kg	141 t	
Regional services	1200	26%	85%	10 kg	119 t	
Coaches	2400	95%	100%	10 kg	355 t	
Rail (<i>SL 20a)</i>						
Railcars	200	100%	100%	18 kg	100 t	
Ships (<i>SL 20a)</i>						
Merchant ships	30	100%	100%	100 kg	36 t	

Source: Other MAC Survey 1999.

- The following Table 15 shows for four selected years from 1995 to 2010 the HFC emissions that follow from the parameters in Table 14. To recapitulate: For MAC systems, generally continuous annual operating emissions of 10% and a final scrapping loss of 30% are assumed.
- In the Business-as-Usual Scenario I, 'other MAC' refrigerant stocks reach 1637 t in 2010 (car A/C by comparison: 28,078 t). More than 90% of this is in road vehicles, i.e. trucks, buses and coaches. Operating emissions amount to 161 t in 2010 (car A/C by comparison: 2792 t) and disposal losses 32 t (car A/C by comparison: 626 t). Both together make up 193 t total emissions in 2010 (car A/C by comparison: 3418 t). Based on the 'other MAC' refrigerant stock, the total emission rate figures 11.8%.

Table 15: 1995-2010 HFC emissions from 'other MAC' applications in t HFC and million t CO₂ equivalent in Scenario I							
	1995 2000 2005 2010						
Operating emissions [t]	13	62	120	161			
Disposal emissions [t]	0	0	10	32			
Total emissions [t]	13	62	129	193			
Ems. in mill. t CO ₂ equivalent 0.02 0.08 0.17 0.25							
Stock [t]	171	675	1247	1637			

Basis: Other MAC Survey 1999. Extrapolation to 2010 based on parameters in Table 14.

• Over the 1995-2010 period, the global warming contribution rises from 0.02 to 0.25 million tonnes CO₂ equivalent (cf. Table 15). This corresponds to five percent of the global warming contribution of emissions from car A/C.

3 2010 Relative Emission Reductions - Scenario II

The operating conditions of MAC systems (vibrations etc.) limit the possibilities to reduce emissions of their refrigerants. Dispensing entirely with air-conditioning may be the most straightforward solution from an environmental perspective, but has been unrealistic since A/C systems became mass-consumed goods in Europe, too. The low-GWP alternate refrigerants currently under debate are above all hydrocarbons (HCs) and carbon dioxide (cf. Reichelt 1996).

3.1 Car A/C Using CO₂ as Refrigerant From 2007 Onwards

- HCs are not expected to have good prospects in car A/C. The risk of explosion through flammable gas penetrating the passenger compartment is generally viewed as too high. The transcritical CO₂ system, in contrast, is viewed by many experts as the next-generation car A/C system (Preisegger 1999).
- The assessment of global warming contribution must not view direct refrigerant emissions in isolation, but must also include the CO₂ emissions of the engine that drives the system (TEWI). In car A/C units the global warming impact of direct refrigerant emissions is of roughly the same order as that of the combustion gases attributable to driving the system (cf. Baker 1999). The energy efficiency of R-134a systems is currently unsurpassed by any other car A/C system. This is why efforts to optimize carbon dioxide systems, whose direct refrigerant emissions are negligible in global warming terms, concentrate on their energy efficiency. The high pressures arising in carbon dioxide systems (130-150 bar in the transcritical range) initially reduced energy efficiency, as they require more robust and thus heavier components. This drawback has been mitigated by means of volume reduction.
- From 1994 to 1997, a joint research programme was conducted by European car manufacturers (BMW, Daimler, VW, Volvo, Rover) and A/C system manufacturers (Behr, Valeo), titled RACE (<u>Refrigeration and Automotive Climate under Environ-</u> mental Aspects). This gave further impetus to the development of carbon dioxide systems. "The RACE group assumes that within 10 years the first series car models with a carbon dioxide system can be introduced to the market" (RACE 1998, 14). It

is anticipated that energy efficiency can reach levels comparable to 134a systems (cf. Köhler/Sonnekalb 1996; Sonnekalb/Köhler 1997; Wertenbach et al. 1997).

- In addition to the Europeans, major Japanese and North American manufacturers and their component suppliers are also working on carbon dioxide systems. Besides costs, the key issue is the worldwide acceptance of the new system. Operating two different A/C technologies in parallel over the long term is not desirable.
- If development continues according to schedule, the first series vehicles equipped with carbon dioxide A/C systems shall be produced between 2005 and 2008. According to VW 1999 and DaimlerChrysler 1999, they could be launched in 2007. This means that in 2010 the operating emissions of R-134a are only slightly lower than in the Business-as-Usual Scenario I, as a conversion period for all models of at least two years is to be expected.
- If carbon dioxide A/C systems are gradually introduced in Germany from 2007 onwards and if 2009 is the first complete annual cohort without R-134a systems, then during the 2007-2010 period only about 3.3 million R-134a systems are installed in newly registered cars instead of 12.5 million. By the end of 2010, 9 million carbon dioxide systems are in operation. Instead of 36.4 million car A/C units à 0.73 kg R-134a, there are in 2010 only 27.2 million R-134a units with a total refrigerant stock of 21,184 t (instead of 28,022 t in Scenario I).



Diagram 9: R-134a operating and disposal emissions from car A/C systems in 1995-2010 in t: Comparison between Scenarios II and I. In Scenario II, after introduction of carbon dioxide systems from 2007 onwards, operating emissions (black area) sink to 2221 t by the year 2010. Disposal emissions remain the same as in Scenario I. The difference in emissions compared to Scenario I (white area at top right) of 571 t is the outcome of the sinking operating emissions in Scenario II, while in Scenario I operating emissions grow, resulting in 3418 t total emissions.

- Operating emissions in 2010 only figure 2221 t instead of 2792 t. This amounts to 571 t or 20% less than in the R-134a Scenario I. The global warming contribution of total emissions (operating plus disposal emissions, the latter being equal in both scenarios) is reduced by 0.74 million t CO₂ equivalent.
- In 2010, the impact of refrigerant substitution is not yet major. From 2021 onwards, however, there are practically no HFC emissions any more from car A/C systems.

3.2 Other Mobile Air-Conditioning

Water chillers in buses from 2000 onwards

- In Germany, as elsewhere, bus manufacturers envisage carbon dioxide as the medium-term option (Mayer 1998). However, a different alternative will be available sooner: packaged 'secondary' A/C systems where chilled water is circulated (water chillers). Beginning in 1999, if customers wish, the first buses are available with only a small A/C unit that chills circulating water, instead of roof-top systems driven by the rear engine that contain more than 10 kg circulating refrigerant (Kirsamer 1999). They can be produced in series from 2000 onwards, and can become the standard in all buses from 2002 onwards. Instead of 10 kg refrigerant, they only require 3 kg R-134a (Schreiber 1999), and the annual operating emission rate is reduced by at least half (5% instead of 10%). If the unit is mounted outside of the passenger compartment, the primary refrigerant loop can also use propane or carbon dioxide (Kirsamer 1999).
- If new bus A/C systems are converted to water chillers (which, it may be noted, cost less than direct R-134a systems) over the 2000-2002 period, then the R-134a stock drops by 2010 to 303 t (instead of 615 in Scenario I). Operating emissions then only figure 16 t (instead of 61 t in Scenario I). Disposal emissions remain unaltered at 10 t. The global warming contribution of the 26 t total emissions is lower by 0.06 million t CO₂ equivalent than that of the 71 t total emissions in Scenario I.

Air cycle systems in railcars from 2000 onwards

Deutsche Bahn AG, the national railway operator, is due to take new high-speed ICE-3 trains into service in the year 2000. These will have air cycle systems (Reum 1999). These systems, which are based on experience made in aircraft air-conditioning (for technology details cf. Adolph 1998), have been undergoing testing in an ICE-1 train since 1995. Their energy requirement is 20-30% higher than that of comparable R-134a systems (Adolph 1999). The decision for air was already taken in 1990, when leakage of the rail A/C systems then in use was causing such high R-12 emissions that their TEWI assessment was 30% worse than the planned air cycle system. R-134a systems have improved in the meantime, so that the air cycle system no longer has a TEWI advantage. It is therefore expected that air cycle systems will remain a transitional episode and will not penetrate the entire railcar stock of Deutsche Bahn AG. Over the long term, carbon dioxide is favoured as the refrigerant of the future in railcars, too (Adolph 1999).

• Of the total R-134a stock of 1637 t in 'other MAC' applications anticipated in 2010 in Scenario I, rail A/C systems only account for 100 t. Through application of air cycle systems and their restriction to the ICE-3, the R-134a stock in 2010 will figure not 100 t, but 90 t. Operating emissions figure 9 instead of 10 t. Savings are small. The one tonne less of emissions reduces the global warming contribution by 1300 t CO_2 equivalent.¹¹

Carbon dioxide in trucks from 2007 onwards

- Due to their technological similarity to car A/C systems, it is assumed in Scenario II that new A/C systems for trucks of all weight classes are also converted to the refrigerant carbon dioxide from 2007 onwards. The R-134a stock of 886 t calculated in Scenario I thus drops to 583 t by 2010. Operating emissions drop from 86 to 61 t, i.e. by 30%. The disposal emissions of 19 t remain equal at first. The global warming contribution is reduced by 0.03 million t CO₂ equivalent.
- If all of the three above abatement measures are implemented namely water chillers in buses, air cycle systems in the ICE-3 high-speed train¹² and carbon dioxide systems in trucks - then in the 'other MAC' sub-sector Scenarios I and II compare as shown in Table 16. The global warming contribution is reduced in the year 2010 by some 71 metric tonnes R-134a or 0.09 million t CO₂ equivalent.

Table 16: R-134a emissions from 'other MAC' applications						
in the years 2000, 2005 and 2010: Comparison between Scenarios I and II*						
2000 2005 2010						
Scenario I emissions [t]	62	129	193			
Scenario II emissions [t] 60 99 122						
R-134a savings in Scen. II [t] 2 30 71						
CO₂ eq. savings in Scen. II [Mt]	0	0.04	0.09			

* Scenario II comprises the abatement measures set out in this Section (2.3.2).

¹¹ An important advantage of air cycle systems deserves mention: They require no storage, disposal and recycling facilities for the refrigerant. ¹² The air cycle system is only assumed for this train, not for others. Other trains will remain with 134a for

the time being.

4 Comparison of Mobile Air-Conditioning Emissions in Scenarios I and II

Due to the dominant role of car A/C systems, the aggregate curves for car A/C plus 'other MAC' are very similar to those already given above for car A/C alone. The changes that result from adding 'other MAC' to car A/C are in the order of 4-6%:

- By adding 'other MAC' to car A/C, the <u>refrigerant stock</u> of all MAC systems is increased by 1637 t (i.e. from 28,022 t to 29,659 t) in Scenario I, and by 1012 t (i.e. from 21,184 t to 22,196 t) in Scenario II. These differences amount to 4% and 5.8%, respectively.
- By adding 'other MAC', <u>annual operating emissions</u> of all MAC systems are in 2010 2953 t instead of 2792 t for car A/C alone in Scenario I, and 2311 t instead of 2221 t in Scenario II. These differences again amount to 4% and 5.8%, respectively.
- The <u>disposal emissions</u> which are identical in both scenarios are 658 t for all MAC as compared to 626 t for car A/C alone. This difference amounts to 4.8%.



Diagram 10: HFC emissions from all MAC systems in t/a: Comparison between Scenarios II and I. Until 2005, emissions in the Business-as-Usual Scenario I and the Reduction Scenario II do not differ. Disposal emissions, setting in from 2003 onwards (grey area at bottom), are identical in both Scenarios. In the 2005-2010 period, operating emissions in Scenario II (black area) no longer rise, this providing a reduction potential (white area) compared to the operating emissions of Scenario I. This potential grows to 642 t by 2010. In 2010, total emissions (operating + disposal emissions) figure 3611 t in Scenario I, and only 2969 t in Scenario II.

- Diagram 10 shows for both Scenarios the development from 1995 to 2010 of total HFC emissions from all mobile air-conditioning.
- In Scenario I, car A/C emissions total approx. 4.44 million t CO₂ equivalent and all MAC emissions 4.69 million t CO₂ equivalent in 2010.
- In Scenario II, car A/C emissions total approx. 3.70 million t. CO₂ equivalent and all MAC emissions 3.86 million t CO₂ equivalent in 2010 (cf. Diagram 11 and Table 17).



Diagram 11: HFC emissions from mobile air-conditioning in million t CO_2 equivalent -Scenarios I and II. The total HFC global warming contributions of the two Scenarios only begin to differ in 2007, when car A/C systems are converted to carbon dioxide refrigerant. By 2010, the difference amounts to 0.83 million t CO_2 equivalent.

• Diagram 11 shows separately for Scenarios I and II how the global warming contribution of emissions from all MAC systems develops over the 1995-2010 period. Table 17 lists the figures for the years 1995, 2000, 2005 and 2010.

Table 17: 1995-2010 HFC emissions from all MAC systems						
in t HFC and million t C	in t HFC and million t CO₂ equivalent in Scenarios I and II					
1995 2000 2005 2010						
Total emissions Scenario I	126 t	1017 t	2352 t	3611 t		
Total emissions Scenario II 126 t 1016 t 2322 t 2969 t						
Ems. in mill. t CO₂ equiv. Scenario I 0.15 1.32 3.06 4.69						
Ems. in mill. t CO₂ equiv. Scenario II	0.15	1.32	3.02	3.86		

3 PU One Component Foam

In can-dispensed polyurethane (PU) one component foam (OCF), Germany is the largest individual market worldwide. Of global sales of some 100 million cans (1997) approx. one quarter is sold here, in can sizes ranging from 300 to 750 ml (Henkel 1999). The arithmetic average foam weight of a can is 660 g, of which approx. 18% is made up by the propellant. In 1997, these propellants were blends, consisting of ca. 40% readily flammable gases (propane, butane, dimethyl ether (DME)) and about 60% HFCs, which are heavier (152a) or nonflammable (134a) (Coens 1999). Since 1995, the HFC proportion in the propellant has been declining. In 1995 it figured 84 g in the 660-g can, in 1997 only 75 g. It is set to drop over the next years to 66 g or 10% of foam weight (Pauls 1999).

The propellant does not serve as a blowing agent or for insulation, but solely to propel the foam from the can. It volatilizes upon application, except for small residues which remain for at most one year in the hardened foam (Kluth 1999). The main application of PU one component foam is to fill cavities and joints when installing inner fixtures in housing construction (AKPU no year). The specific type of window construction commonly practised in Germany favours high levels of consumption (Rathor 1999).

80% of the approx. 24 million cans sold in 1997 in Germany were imported. Their HFC propellants were composed one third of 152a and two thirds of 134a. In the approx. 16 million PU foam cans filled in Germany, of which some 75% are exported, the proportion of 134a is higher at 80-90%.

1 1995-1997 Consumption and Emissions in Germany

The cans are used by professional craftsmen or DIYers half a year after purchase at the latest. HFC emissions of the year n are therefore defined as the sum of half of the sales of the year n-1 and half of the sales of the year n. To this are added the filling losses in manufacture in Germany in the year n, which amount to approx. 0.5 g per can (Henkel 1999). Spent cans are not entirely empty upon disposal, but still contain about 10% of the foam, including its propellant (Jungbluth 1999). The greater part of this propellant is released to the atmosphere, with a time delay. This also applies to the 30% of the cans which are consigned to the central recovery facility for PU foam cans in Thurnau, Germany (Schwarz/Leisewitz 1996, 113).

1995-1997 annual emissions from use and (this being the smaller proportion) from filling are shown in Table 18.

Table 18: 1995-1997 emissions of HFC propellants for PU one component				
foam, in t HFC and million t CO ₂ equivalent				
	1995	1996	1997	
152a + 134a [t HFC]	1680	1749	1805	
Mill. t CO₂ equivalent	1.36	1.39	1.48	
Can sales in millions	20	22	24	

Source: Interviews with major PU can foam fillers and dealers

2 2010 Business-as-Usual Emissions Forecast - Scenario I

Sales of PU one component foam cans in Germany have risen by 2 million cans annually over the 1994-1998 period - from 18 million in 1994 to 26 million in 1998 (Rathor 1999). Sales are expected to continue to rise.

Assumptions for Scenario I:

- 1. Until 2010, can sales in Germany grow annually by 5%.
- 2. The German market grows exclusively through imports, while German production remains at 16 million cans per year.
- 3. The propellant ratio between 152a and 134a remains at 1 to 2 for imported cans, and at 1 to 7 for cans filled in Germany.
- 4. The HFC share in foam weight drops steadily from 75 g to 66 g per 660-g normal can.

This means that in the year 2009 approx. 44 million and in the year 2010 approx. 46 million cans are sold in Germany, each containing 66 g HFC propellant. This results in emissions in the use phase of just under 3000 t in 2010. This is joined by 8 t losses in filling in Germany. Total emissions in 2010 figure 1000 t 152a and 2000 t 134a, corresponding to 2.74 million t CO_2 equivalent.



Diagram 12: 1995-2010 HFC emissions from PU one component foam cans in tonnes. Until 2000, total emissions of 134a (2/3) and 152a (1/3) rise to just under 2000 t/a. In business-as-usual Scenario I, they continue to rise until the year 2010, reaching 3000 t. In Scenario II, propellants are converted to halogen-free hydrocarbons by the year 2010 (apart from 5% 134a). Annual 134a emissions then only amount to 150 t. (See following text.)

3 2010 Relative Emission Reductions - Scenario II

- In Central Europe, the relative HFC quantity per can is regulated since 1993 by a voluntary agreement of the working group of leading Central and Western European fillers (AKPU no year). The '50 g rule' established by this agreement provides for a maximum per 750-ml can of 50 g flammable gases (propane, butane, DME), in order to prevent an explosive atmosphere from arising when a whole can is emptied in a confined closed room (1.56 m³) (Cocon 1996). Through blending with 134a or 152a, this concentration is prevented. 152a is flammable, but only at a concentration roughly 3 times higher than hydrocarbons, so that the blend is held below the lower explosive limit (LEL).
- In Northern Europe, the flammable gases propane/butane/DME are used as propellants without halogens. Because of the cost reductions that this offers, Western and Central European fillers also dispense with HFC in the propellant blend for supplies to Eastern Europe. In the USA (approx. 15% of the global market) only about one third of OCF is propelled with HFCs - mainly by fillers based in Europe. One third uses propane/butane, and the other third HCFC-22. While in Europe only cans < 750 ml are used, in the USA containers exceeding 50 litres are also used.
- In countries without HFC propellants, such as in Scandinavia, no accident attributable to the flammability of the propellants has occurred since HFCs were dispensed with. No cases of deflagration are known in Eastern Europe, either.
- As an alternative to the propellant 134a, there are two options:

• While maintaining the 50-g rule, conversion to 152a, whose GWP is only 140 instead of 1300 (134a). Use of HFC-134a only for safety reasons in small closed rooms and underground (market share 5%).

• Departure from the 50-gram rule and phase-out of HFC in favour of propane, butane and DME. Use of HFC-134a only for safety reasons in small closed rooms and underground (market share 5%).

- The first option is already being used today by two of the ten largest European fillers. It is not only realizable but is already being realized.
- The second option is already practised for the markets outside of Central Europe. Several of the ten largest European fillers count among the companies using flammable hydrocarbons instead of HFCs for these markets. (The ten largest European fillers account for 80% of global sales.)
- A debate is under way among the ten largest European OCF fillers on the continuation for Central Europe of the restriction to 50 g flammable propellants per 750-ml can. The explosions feared in use of HFC-free OCF cans have not occurred (Henkel 1999). The arguments in favour of HFCs are losing weight. The main barrier to the introduction of more or exclusively halogen-free propellants is a lack of explosion protection in existing filling facilities.

- From the climate protection perspective, conversion of OCF propellants from 134a to 152a is a good solution, while conversion from HFCs to halogen-free hydrocarbons is an even better solution. It is assumed in Scenario II that by the year 2010 HFC propellants are in principle substituted by hydrocarbons. In this Scenario, HFC propellants are only used for applications in confined closed rooms and underground, accounting for approx. 5% of the total market.
- As a result, emissions in 2010 in Scenario II are only 150 t (exclusively 134a) instead of 3000 t HFCs. This is a reduction of 95% compared to Scenario I.
- This means emissions in 2010 of only 0.19 million tonnes CO₂ equivalent instead of 2.74 million tonnes. This is a reduction of global warming impact by 93% (cf. Diagram 13).



Diagram 13: 1995-2010 HFC emissions from PU one component foam (OCF) cans in million t CO_2 equivalent - Scenarios I and II. From 1995 to 2000, development follows Scenario I, in which the global warming contribution of OCF propellants rises by the year 2010 to 2.74 million t CO_2 equivalent. If, however, HFCs are successively left out of propellant blends from the year 2000 onwards, the global warming contribution is cut to 0.19 million t CO_2 equivalent.

Table 19: 1995-2010 global warming contribution of PU one component foam							
propellants, in million t CO ₂ equivalent							
	1995 2000 2005 2010						
Scenario I	1.36	1.74	2.17	2.74			
Scenario II	1.36	1.74	0.96	0.19			

Note: Real emissions in 1995, from then onwards forecasts

4 Products of PU Foam

PU foam products consist of flexible foam, integral skin foam and rigid foam. Flexible foam requires neither HCFCs nor HFCs as blowing agents. In rigid foam, HCFCs are now used as blowing agents, and HFCs very recently since 1998. In the 1995-1997 period, HFCs were only used in integral skin foam.

1 1995-1997 Consumption and Emissions in Germany

The main application of integral skin foam, in which, since 1996, small quantities (1%) of HFC-134a have been added as auxiliary blowing agent in order to improve product quality, is the manufacture of soles for sports and leisure shoes. Chemically speaking, this is because the polyols (reaction components of isocyanate) in this application are not polyester polyols, but polyether polyols.

HFC consumption in Germany for integral skin foam products figured some 60 t in 1996 and 70 t in 1997. The HFCs do not serve thermal isolation. They do not remain in the product, but are emitted in the year of production. Consumption and emission levels are thus equal.

Table 20: 1995-1997 HFC consumption/emission for integral skin foam,						
in t HFC and million t CO ₂ equivalent						
1995 1996 1997						
HFC-134a [t]	0	60	70			
Ems. in million t CO_2 equivalent00.080.09						

Sources: Wiedermann 1999; Kroesen 1999; Schubert 1999.

At present (1999), consumption is continuing to rise. The experts interviewed expect it to remain at a constant level of 120 t/a from the year 2000 onwards. Replacement of 134a - e.g. by liquid HFCs - is not under consideration over the medium term.

2 Present and Future PU Rigid Foam Blowing Agents

It is possible for PU rigid foam to be blown exclusively by CO₂ generated by the chemical reaction of water, which is contained in small quantities in the polyol formulation, with the isocyanate. The blowing agent CO₂ generally diffuses very rapidly from the foam cells that it forms. It is replaced by ambient air, which reduces insulation performance. This then needs to be compensated for by greater foam thicknesses. This technique is adequate for applications where the insulating effect is secondary to other goals (such as filling cavities).

Where good thermal insulation (low thermal conductivity) is essential, a physical blowing agent is added to the 'chemical blowing agent' CO_2 formed spontaneously upon foaming. This physical agent remains in the foam cells. Instead of the physical blowing agent CFC-11, which was formerly used universally, HCFC 141b - which is also liquid and hardly combustible at room temperature - is now used worldwide, sometimes also the HCFC blend 22/142b. The quantities applied amount to approx. 9% of the foam weight. HCFCs will be banned in a few years in the industrialized countries.

2.1 HFC-134a as Co-Blowing Agent for Sandwich Panels

The three largest applications of PU rigid foam, with 50,000 t annual foam production each, are, in Germany: <u>firstly</u> domestic refrigeration, <u>secondly</u> flexibly faced (e.g. with aluminium foil) insulating panels for roof insulation, and <u>thirdly</u> sandwich panels, i.e. insulating slabs faced rigidly (with steel plate) as structural components for factory buildings, cold stores etc. In contrast to many other countries, in Germany the blowing foam most commonly used for PU rigid foam is not an HCFC, but the halogen-free hydrocarbon pentane. In domestic appliances, pentane is used exclusively as the blowing agent, in flexibly faced continuous panels it is used in over 90% of production (IVPU 1999). As do HCFCs, pentane remains in the foam cells and thus contributes to thermal insulation. However, its use necessitates explosion-proof processing facilities, new tanks and piping etc. This means high investments, which only pay back through the low blowing agent price with larger throughputs.

Sandwich panels, in contrast, were still blown predominantly with HCFCs in 1997. Since 1998, a conversion process is under way which will lead by the year 2000 to half of the foam also being blown with pentane and the other half predominantly with CO_2 complemented by the addition of 134a as a co-blowing agent (Sandwich Panel Survey). The 134a remaining in the foam cells permits a lower density of the foam (weight savings!) for a given insulation performance: its specific insulation performance is improved. Moreover, this prevents shrinking of the foam core caused by rapid CO_2 diffusion. Due to low solubility of HFC-134a in the polyol component, its use vis-à-vis HCFC-141b is limited to approx. one third (3% of foam weight) (Schubert 1999).

As HFC-134a has only played a role since 1998 as physical co-blowing agent for CO_2 (water) blown sandwich panels, it does not yet figure in the 1995-97 statistics for PU rigid foam, but is included in the emissions forecast for 2010.

2.2 New Liquid HFCs for Small-Scale Applications

There are a number of further applications of PU rigid foam that are usually aggregated as 'others', and currently use HCFC-141b, which will become relevant in the future for HFC blowing agents (Jeffs/de Vos 1999). These include spray foam for roof insulation, whose open in-situ application requires hardly flammable liquid blowing agents for safety reasons. Pentane cannot be used here. <u>Discontinuous</u> insulating foams have greater quantitative importance: discontinuous block foam from which, for instance, insulating moulds are cut, or small-series insulating panels for refrigerated vehicles, cold storage cells, commercial refrigeration etc. A part of the continuous panels with flexible facing is also not blown with pentane, but with 141b. These applications are characterized firstly by high requirements upon insulation performance which can only be achieved by means of blowing agents that remain in the foam cells, and secondly by small- to medium-sized processors for whom conversion from 141b to pentane is too expensive (Wiedermann 1999; Szily 1999; Esser 1999).

In the EU, Japan and the USA, 141b will be banned as a foam blowing agent from 2003/2004 onwards. The chemical industry intends to make new liquid HFC-based blowing agents commercially available by that date. These are the HFCs 245fa and 365mfc, with boiling points of 15.3°C and 40.2°C, respectively (UNEP-FTOC 1998).

245fa and 365mfc are treated here as a uniform substance because their respective consumption levels can not yet be anticipated today and their GWP values scarcely differ: 820 for 245fa, 810 for 365mfc. The aggregate GWP for '245fa/365mfc' is 815.

2.3 Emission Profiles of 134a and 245fa/365mfc From PU Rigid Foam

The production process of foams entails high blowing agent emissions, while annual losses from the finished product are low. The emission profile is characterized by high first year losses (10 to 25% of the quantity applied), while annual losses in the use phase until decommissioning only amount to 0.5 to 1.5%. Table 21 compiles the standard values for various 134a and 245fa/365mfc applications according to IPCC recommendations.

Table 21: Emission identification PU rigid foam					
HFC-134a applications	Product life	First year loss	Annual loss		
	[years]	[%]	[%]		
PU - Continuous panel	50	10	0.5		
HFC-245fa/365mfc applications					
PU - Discontinuous panel	50	10	0.5		
PU - Discontinuous block	50	10	1		
PU - Spray	50	25	1.5		
PU - Injected	15	10	0.5		
PU - Continuous panel	50	7.5	0.5		

Source: IPCC 1999, Appendix 2 (p. 64).

First year losses are the emissions during the foaming process (plant losses). For 134a in sandwich panels (continuous panels) these are estimated at 10% of the quantity applied. Annual losses in use (stock emissions) are estimated at 0.5%. With these figures, 65% of the blowing agent are still contained in the foam upon decommissioning after a 50-year product life. What happens with this remainder is not yet relevant by the year 2010. This applies similarly to 245fa/365mfc applications. Here the production loss (plant losses) averages 12.5% and annual losses during use in the stock average 0.8%.

3 2010 Business-as-Usual Emissions Forecast - Scenario I

The forecast for 134a as a co-blowing agent assumes firstly that usage of integral skin foam remains constant at 120 t/a and, secondly, that from 2000 onwards half of the sandwich panel rigid foam is blown with pentane and the other half with CO_2 (water) plus 134a. These only use 3% HFC-134a in relation to foam weight, instead of some 9% HCFC (141b or 142b/22). If 25,000 t rigid foam are produced in this manner per year (here: the year 2000), then the quantity of 134a applied amounts to some 750 t. Given an annual growth rate for sandwich rigid foam of 5%, the quantity applied rises to 1220 t by the year 2010. (This is joined by a constant 120 t HFC-134a for integral skin foam products.)

Industry circles expect a maximum annual requirement in Germany for the new liquid HFCs 245fa or 365mfc of approx. 2500 t/a in the year 2010 (VCI 1999; Wiedermann 1999). This estimate is based on two preconditions. Firstly, 'other' foam production grows annually by 5% until 2010, to a level of 30,000 t/a. Secondly, the present market share of halogenated blowing agents remains constant, HCFC-141b being replaced by the new liquid HFCs. With these assumptions, consumption in the first full year of use (2004) can figure 1850 to 1900 t, and may then rise annually by 5% up to 2500 t/a. The quantity of liquid HFC applied is 8 to 9% of foam weight.

3.1 134a Emissions Until 2010

134a emissions from German production (plant emissions) rise over the 2000-2010 period from 75 to 122 t without and from 195 to 242 t with integral skin foam. Annual losses from the German rigid foam stock (the German market is smaller by one third than production in Germany) are still low in the first years of product life. They grow over the 2000-2010 period from just under 2 t to 29 t. Total emissions (including integral skin foam) grow from 197 to 272 t. This is a growth in the global warming contribution from 0.26 to 0.35 million t CO_2 equivalent.

3.2 245fa/365mfc Emissions

The plant emissions of liquid HFC blowing agents, which are only introduced fully by the year 2004, are higher than those of 134a. This is because their share of foam weight is higher - 8-9% instead of 3%. Plant emissions figure 233 t in 2004, and 313 t in 2010. The annual emissions from the foam product stock in these two years are 7 and 94 t (in these applications, the size of the German market is roughly the same as that of production in Germany). Total emissions rise from 240 to 407 t. This is a growth in the global warming contribution from 0.2 to 0.33 million t CO_2 equivalent.

4 2010 Relative Emission Reductions - Scenario II

The reduction scenario assumes that after HCFC phase-out in rigid foam the movement towards zero-GWP substances continues. Even without statutory pressures, the blowing agent whose application appears more secure over the long term will establish itself for preventive reasons, wherever that blowing agent is not detrimental to insulation performance.

In sandwich panels, major manufacturers who are using HFC-134a as a co-blowing agent with CO_2 since 1998 plan to shortly dispense with the HFC again in the greater part of their product range. This shall be made possible by, among other things, improvements in the cell structure of CO_2 -blown rigid foams (Sandwich Panel Survey).

Many manufacturers in 'other' PU rigid foam applications who continue to use HCFC-141b, which is still permitted at least until 2003, have the choice when converting between pentane, CO_2 (water) and the new HFC technologies. The above-mentioned fears concerning the long-term security of supply of global warming substances can be expected to favour pentane and CO_2 (water) here, too, particularly as uncertainties continue to prevail concerning the prices and commercial availability of liquid HFCs.

4.1 HFC Usage Reduced by 50% by 2010 Compared to Scenario I

Scenario II assumes for sandwich panels that a departure from 134a as co-blowing agent already sets in from 2001 onwards, resulting in the quantities applied being reduced by half by the year 2010 compared to Scenario I - from 50% of cases to 25%.

For the 'other' applications, Scenario II assumes that in 2004 not all but only 90% of previous 141b users switch to liquid HFC and that their proportion drops to 50% by the year 2010 compared to Scenario I.

For integral skin foam, no differences to Scenario I are assumed.

4.2 Reduced HFC Emissions

In rigid foam, manufacturing plant emissions of <u>134a</u>, starting at 75 t in the year 2000, do not rise to 122 t in 2010 but rather drop to 61 t. Annual losses in use do not rise to 29 t (Scenario I) but only to 22 t. Total emissions in 2010 figure just under 83 instead of 152 t (cf. Diagram 14). This corresponds to a global warming contribution that is reduced compared to Scenario I by 90,000 t CO_2 equivalent.



Diagram 14: 1998-2010 HFC-134a emissions from PU rigid foam in t/a - Scenarios I and II. In Scenario I, 134a is used as co-blowing agent in 50% of PU sandwich panels, in Scenario II only in 25% by the year 2010. Total emissions in 2010 figure 83 t in Scenario II (lower area) instead of 152 t in Scenario I (total of both areas).

The plant emissions of <u>liquid HFCs</u> in the year 2010 do not figure 313 t, but only 156 t. Annual losses in use do not rise from 7 t (2004) to 94 t, but only to 67 t in 2010. Total emissions in 2010 figure 223 t instead of 407 t (cf. Diagram 15). For liquid HFCs, Scenario II thus reduces the global warming contribution by 150,000 t CO_2 equivalent compared to Scenario I.



Diagram 15: 2002-2010 HFC 245fa/365mfc emissions from PU rigid foam in t/a - Scenarios I and II. In Scenario I, HCFC-141b is replaced from 2004 onwards entirely by liquid HFCs. In Scenario II replacement is not complete, but only takes place in 50% of cases by 2010. Total emissions in 2010 figure only 223 t in Scenario II (lower area) instead of 407 t in Scenario I (total of both areas).

If HFC-134a emissions from integral skin foam blowing, which are equal in both scenarios, are taken into consideration, we receive for PU foam products the following HFC emissions over the 2000-2010 period in the two scenarios (Table 22).

Table 22: 2000-2010 HFC emissions from PU foam products - Comparison of						
Scenarios I and II						
2000 2005 2010						
Scenario I emissions [t]	197	495	679			
Scenario II emissions [t] 197 413 426						
Emissions savings in II [t] 0 82 271						
CO ₂ equiv. savings in II*	0	0.08	0.23			

* million tonnes CO₂ equivalent

5 Global Warming Contribution of all PU Rigid Foam HFCs - Scenarios I and II

As shown in Diagram 16, 134a and 245fa/365mfc emissions in the year 2010 amount to 0.68 million t CO_2 equivalent in Scenario I and 0.45 million t in Scenario II. These are the emissions from integral skin foam and PU rigid foam in small-scale applications and in sandwich panels.



Diagram 16: 1995-2010 HFC emissions from PU foam products in million t CO_2 equivalent. In Scenario I the global warming contribution rises until 2010 to 0.68, in Scenario II to 0.45 million t CO_2 equivalent.

5 XPS Rigid Foam

Foam panels made of extruded polystyrene (XPS) have the advantage over PU panels of being more water-repellent and extremely compression-resistant. They are used mainly for exterior insulation in building construction, and are suitable for moist areas (exterior walls in contact with the soil). For XPS panels, too, there was no consumption or emissions of HFCs prior to 1999. From then on, however, very large quantities are expected.

German manufacturers already replaced CFC-12 as blowing agent and cell gas by HCFCs in 1990, namely either by 142b or by a mixture of 142b and 22. In 1996, the two most important producers in Germany, BASF and Dow Chemical, entered into a voluntary agreement to dispense with HCFCs from 2000 onwards. It is thus not expected that the producers switch from the 142b/22 mixture, which will be banned from 2000 onwards, to pure 142b, which will continue to be permitted for a further four years. Instead, all four German producers are working on systems using zero-ODP substances, which will be used entirely from 2000 onwards (XPS Foam Survey).

1 Blowing Agents From 2000 Onwards

Most German producers are orienting themselves to HFC systems. The blowing agents coming into question here are 152a and 134a, either alone or mixed with each other.

<u>152a:</u> The advantage of 152a is that it has very good solubility in polystyrene, thus producing a very fine and homogeneous cell structure. Its disadvantage is the rapid and total diffusion from the foam cells, which takes place within only a few weeks¹³. This compromises insulation performance, meaning that 152a, if used alone, is not suitable for the entire product mix. Moreover, it is flammable, requiring expenditure for production safety.

<u>134a</u>: The nonflammable HFC-134a has the advantage of remaining in the finished foam and thus guaranteeing good insulation performance. However, in contrast to 152a, it has poor solubility in polystyrene, so that it is unable to produce a cell structure suited to a full product mix. 134a will therefore be used to improve thermal insulation in combination with another blowing agent, being either 152a or CO_2 .

<u>CO₂</u>: As in PU rigid foam, CO₂ is a candidate blowing agent. However, in XPS it is not formed by a chemical reaction but must be provided externally. With CO₂ alone, only a small part of the product range can at present be produced without loss of insulation performance. Among the panel thicknesses ranging up to 200 mm, only those with thicknesses of less than 60-70 mm have a quality comparable to those foamed with HCFCs. However, when applied in combination with 2-3% organic blowing agent, namely ethanol, the entire product mix can be produced without loss of quality - as stated by the developer of this process, BASF (Boy 1999). However, production facilities need to be structurally modified for the flammable blowing agent, which entails high capital investment.

¹³ IPCC literature states for 152a-blown XPS foam a first year loss of only 40%. This value is viewed by experts as too low (Abbott 1999; Schubert 1999).

German producers are in agreement that high-quality XPS panels can be produced with 152a and 134a. Nonetheless, BASF views HFCs only as a transitional solution and is currently planning to convert its production in Germany from 2000 onwards entirely to CO_2 or CO_2 plus ethanol. However, if market acceptance is lacking, BASF states that it would see itself forced to use HFCs (Boy 1999).

Consumption and Emission Factors

The input of halogenated blowing agent per cubic metre foam figures 4-5 kg. Of this, 15 to 25% are already emitted in the production facility (UNEP-FTOC 1998, xvii), while other sources report 25% (Ashford 1999) or 30% (Boy 1999). This high loss is an outcome of the largely open foaming process. The foaming agent is injected and dissolved under high pressure in an extruder with molten polystyrene granulate. When the melting exits the extruder to the chilled foam band, the blowing agent vaporizes due to the pressure drop, thus foaming the polystyrene (Boy 1997). Capture and recovery have been tested - but for technical and economic reasons not yet implemented (UNEP-FTOC 98, xvii).

If 152a is used as blowing agent, the 152a remaining in the product already largely diffuses out of the panels in plant storage (Schubert 1999). 134a, in contrast, remains in the product and diffuses almost as slowly as 142b. The half life in a 100 mm thick panel is 76 years for 134a and 84 years for 142b (Weilbacher 1987). The annual emission rate of 0.66% for the entire product mix assumed here is based on 100 mm thick panels. Table 23 lists the emission factors applied here.

Table 23: Emissions identification for XPS foam						
Blowing agent Product life First year loss Annual loss						
	[years]	[%]	[%]			
HFC-134a	50	25	0.66			
HFC-152a	50	100	-			

Sources: Boy 1997; Weilbacher 1987. The IPCC values (IPCC 1999) for 152a (40% first year loss) were not adopted.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

Scenario I assumes the - conceivable - case that <u>all</u> German producers use the HFCs 152a and 134a as blowing agents. A ratio of 1 to 1 between the two HFC blowing agents is assumed, not for individual products or producers but for the entire product mix produced in Germany.

In 1998, 1.1 million cubic metres of XPS insulation material were sold in Germany (GDI 1999). With a net export surplus of 30-40%, German production amounted to about 1.5 million m³. An annual production growth of 5% is anticipated until 2010 (Abbott 1999), so that German production can then be expected to amount to 2.7 million m³ and the German sales market to 2 million m³.

Under these circumstances, the emissions of 134a and 152a produced in Germany rise over the 2000-2010 period from 4650 t to 7575 t. This large quantity is an outcome of the circumstance that 152a is emitted completely in the first year, and 25% of the 134a

is released in the year of production (Table 23). The 7575 t emissions in the year 2010 are composed of 6060 t 152a (i.e. 80%) and only 1515 t 134a. However, the global warming contribution of 134a is more than twice (2.3-fold) that of 152a, the latter having a relatively low GWP (140).

Annual losses in the use phase from the XPS foam stock in Germany (the German market is smaller by a quarter than German production) consist only of 134a. Over the 2000-2010 period, they rise from just under 3 t to 169 t. In the year 2010, too, total emissions are dominated in quantitative terms (almost 98%) by plant emissions, and then amount to 7745 t. The global warming contribution of total emissions rises over the 2000-2010 period from 1.73 to 3.04 million t CO_2 equivalent.

3 2010 Relative Emission Reductions - Scenario II

Scenario II assumes that from the year 2000 onwards the greater part of XPS insulation foam in Germany is produced with the HFCs 152a/134a. The smaller part (43%) is produced using CO_2 and ethanol. The share of 43% foam without HFCs in an overall production volume growing annually by 5% is held constant until 2010, although it is to be expected that if halogen-free foamed XPS foam gains sufficient market acceptance this process will be used increasingly in the subsequent years. Insofar, Scenario II is based on conservative assumptions.



Diagram 17: 1999-2010 HFC 152a/134a emissions from XPS foam in t - Scenarios I and II. In Scenario I, HFCs are used exclusively for foaming, their total emissions (predominantly 152a) rising over the 2000-2010 period to 7745 t (total of both areas). In Scenario II, 43% of production is without HFCs, so that their emissions grow until 2010 to only 4415 t (dark area), offering a corresponding reduction potential. Until 2000, HCFCs are used as blowing agent.

Under these conditions, plant emissions of 134a and 152a in Germany commence in the year 2000 not at 4651 t but at 2651 t. In the year 2010, they do not figure 7575 t (Scenario I), but 4318 t (cf. Table 24).

The annual 134a losses through emissions from the foam stock in use rise until 2010 not to 169 t but only to 96 t.

Total emissions figure in 2010 only 4415 t (3455 t 152a + 960 t 134a) instead of 7745 t (Scenario I). Cf. also Diagram 17.

Table 24: 2000-2010 HFC emissions from XPS foam in Scenarios I and II in tonnes									
	2000 2005 2010								
	Scen. I	n. I Scen. II Scen. I Scen. II Scen. I Scen. I							
Plant emissions	4651	2651	5936	3384	7576	4318			
Stock emissions	3	2	77	44	169	96			
Total emissions 4654 2653 6013 3427 7745 4415									

4 Comparison of Global Warming Contributions of Scenarios I and II

In the year 2010, the global warming contribution of total emissions in Scenario II figures 1.73 million t CO_2 equivalent. The 43% reduction compared to the 3.04 million t of Scenario I (cf. Diagram 18) yields the considerable difference of 1.3 million t CO_2 equivalent.



Diagram 18: 1995-2010 HFC emissions from XPS rigid foam in million t CO_2 equivalent (Scenarios I and II). The global warming contribution of total emissions is generally 43% lower in Scenario II than in Scenario I; in the latter, the total global warming contribution in 2010 is approx. 3.04 million t CO_2 equivalent.

6 Aerosols: Metered Dose Inhalers

Under the exemptions for essential uses under the Montreal Protocol ozone regime, CFCs are presently still available in Europe for metered dose inhalers (MDIs). These are permitted for the treatment of asthma and chronic obstructive respiratory diseases as long as equivalent alternatives are not sufficiently available. Reformulating the numerous medications with HFCs instead of CFCs requires considerable research and expenditure. In Germany, the first MDI using the propellant 134a came onto the market in April 1996, followed in 1997 and 1998 by two MDI products in each year, and one in 1999 (all using 134a). These six products of two British manufacturers were the only CFC-free MDIs on the German market by the middle of 1999.

1 1995-1997 Consumption and Emissions in Germany

Table 25: 1995-1998* HFC emissions from metered dose inhalers								
in t HFC and million t CO ₂ equivalent								
1995 1996 1997 1998*								
134a emissions [t] 0 0.25 9 26.7								
Ems. in million t \overline{CO}_2 equiv. 0 0.000 0.012 0.035								

Source: Inhaled Drug Delivery System Survey. The figures are sales in pharmacies, not factory production. A time lag, such as for PU one component foam cans, does not need to be taken into consideration here for the emissions assessment. * Preliminary value for 1998.

As shown in Table 25 and Diagram 19, HFC emissions from the use of MDIs were still very low over the 1995-1997 period: 250 kg in 1996 and 9 t in 1997. Their global warming contribution (as yet only 134a) was accordingly low until 1997. Over the same period, some 400 t CFCs were released annually from the more than 24 million MDIs in use in Germany. Only in 1998 - according to preliminary data - did the latter figure drop by approx. 7%, because some 27 t of the propellants were then HFCs.



Diagram 19: 1995-1998 HFC emissions from MDIs in t HFC. Due to the high market share of CFC-propelled medications, HFC emissions are still low but exhibit a steep rising tendency.

It is expected that the European Commission proposal (EU-COM 98) presented in late 1998, which was recently approved by the environment committee of the German Bundestag (parliament), will accelerate the transition from CFCs to HFCs. Under this proposal, the annually adopted exemption for CFC-propelled MDIs shall be refused if sufficient quantities of HFC-propelled alternatives are available for the same medication. The use of CFCs could expire by the year 2003 for all classes of medication.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

- Inhaled drug delivery systems are standard in the therapy of asthma and COPD (chronic obstructive pulmonary disease). In many cases, the patient must regularly apply two substances, one to dilate the bronchi and one to control inflammation. Propellant-containing MDIs ('sprays') are not the only form of inhalation, but in Germany the most important one. Scenario I assumes that this continues to be the case. This means that the 400 t CFC consumption in Germany in 1995 is the standard by which to estimate the quantity of HFCs emitted annually after complete CFC phase-out.
- In view of the further growth in prevalence of asthma and COPD predicted by all sides, an equal tonnage of propellants, which account for 99% of the content of MDIs, may appear too small in the year 2010. However, two points need to be considered in this connection. Firstly, with the same filling volume an HFC-propellant is lighter than a CFC-propellant (1.20 as compared to 1.38 density).¹⁴ Secondly, for the same number of doses, the HFC cans presently on the market are somewhat smaller than their CFC-propelled predecessors. The two factors combined make it plausible to assume for Scenario I that the propellant quantity remains constant by weight at 400 t/a, despite the increased use of inhalation therapy.
- In addition to 134a, the pharmaceutical industry has also tested HFC-227 and has found it suitable. The decision for 134a or 227 depends mainly upon the chemicalphysical compatibility with the active ingredient to be formulated. In the present study, a 75:25 ratio of 134a:227 usage is assumed for the entire medication mix (the GWP of 227 is 2900).
- From the above, emissions of 300 t 134a and 100 t 227 follow for both the years 2005 and 2010 in Scenario I. In both of these years, the global warming contribution of the two HFCs figures 0.68 million t CO₂ equivalent.

3 2010 Relative Emission Reductions - Scenario II

• In the German market for inhalation delivery systems, propellant MDIs have a share of 75-80%. Their success is due to the circumstance that for decades they were the only technology capable of delivering 100 or 200 exactly measured doses, in a simple and discrete manner. There have always also been - propellant-free - dry powder inhalers (DPIs). These, however, were complicated to handle and in many cases only delivered a few or often even only a single dose of the active ingredient. They were intended for the minority of patients who did not tolerate sprays.

¹⁴ CFC-propelled MDIs had a content of 5 to 30 ml in 1997 - a weighted average of 12 ml or 16.5 g.

- The vigorous search that set in at the end of the 1980s to find CFC-free but equivalent processes led to an international 'innovation thrust' (Matthys 1995), and resulted in optimized DPI systems. By 1996, three pharmaceutical companies had marketed in Germany new DPIs which deliver 60 or 200 exact individual doses just like MDIs, and are just as simple and discrete in operation.
- By 1998, the debate on this issue among doctors, pharmacists, pharmaceutical companies and environmental associations had arrived at the conclusion that modern DPIs are of the same medical value as MDIs. Deutsche Atemwegsliga, the association of leading pneumologists, has declared for active ingredients not intended for acute attacks that "DPIs are a safe and convenient form of medication and are preferable to CFC-propelled MDIs" (Wettengel 1998). MDIs cannot be dispensed with completely: small children, patients with acute asthma attacks or with severest respiratory diseases do not have the strength to draw sufficient powder into their respiratory tract. The group of patients dependent upon MDIs is estimated by experts at only "a few percent" (Schultze-Werninghaus 1996, 20).
- Scandinavia has shown that asthma and COPD can be treated well with modern DPIs. In these countries, DPIs have a market share of 75%. The German DPI working group (Arbeitskreis Pulverinhalation, API) aims at "Scandinavian conditions" for Germany, i.e. a DPI share of 75 instead of 25 percent (API 1999). In addition to doctors, pharmacists, patients and environmental associations, this working group includes since 1997 the GlaxoWellcome, AstraZeneca and Orion Pharma companies, leading pharmaceutical companies in the field of respiratory therapy worldwide, all of whom concentrate their German sales on DPIs.
- Scenario II implements the targets of this working group, which aims to limit over the medium term the market share in Germany of inhalation delivery systems using halogenated propellants to a medically indicated ceiling of 25%. In their place, the share of propellant-free DPIs is to rise from currently 20% to 75%.¹⁵

Under these conditions, HFC emissions in 2010 do not figure 400 t, but only 100 t (with a 134a:227 ratio of 75:25). As shown in Table 26 and Diagram 20, HFC emissions in the year 2005 are higher. The temporary rise to 200 t is possible because for numerous medications presently on the German market only HFC-propelled MDIs are planned as replacements for CFC-propelled systems.

Table 26: HFC emissions from MDIs in 2000, 2005 and 2010:							
Comparison of Scenarios I and II							
2000 2005 2010							
Scenario I emissions [t HFC]	120	400	400				
Scenario II emissions [t HFC]	120	200	100				
Scenario II savings [t HFC]	0	200	300				
Scen. II savings [million t CO ₂ equiv.]	0	0.340	0.510				

¹⁵ Boehringer Ingelheim, the leading pharmaceutical company in Germany in the field of asthma medication, has not developed a multi-dose DPI, but is working on a propellant-free 'pocket nebulizer' - a type of micro-air-jet pump (Inhaled Drug Delivery System Survey). IG Sprühtechnik, an aerosol filling company, is testing isobutane as propellant for MDIs (Guck 1998). Both of these technologies would contribute to replacing HFC systems as in Scenario II. However, neither of the two is considered to have the same medium-term prospects as DPI. Wright 1999 gives an overview of potential future inhalation technologies.



Diagram 20: 1996-2010 HFC emissions from MDIs in t HFC - Scenarios I and II. In Scenario II, HFC emissions figure not 400 in the year 2010 as in Scenario I, but only 100 t, because by then DPIs have replaced three quarters of the MDI market.

The global warming contribution (cf. Diagram 21) in 2010 figures only 0.170 million t CO_2 equivalent in Scenario II compared to 0.680 million t in Scenario I.



Diagram 21: Global warming contribution (million t CO_2 equivalent) of HFC emissions from MDIs. The global warming contribution in 2010 figures only 0.170 million t CO_2 equivalent in Scenario II compared to 0.680 t in Scenario I.

7 Semiconductor Manufacturing

In the semiconductor industry, HFC-23 (i.e. CHF_3) is an important gas for dry etching in low-pressure plasma. CHF_3 is treated in Part IV of this study together with other fluorinated gases used in semiconductor manufacturing. The following data on consumption and emissions are taken from Part IV.

1 1995-1997 Consumption and Emissions in Germany

Table 27: 1995-1997 HFC-23 consumption and emissions in the semiconductor industry [t/a]								
	1995 1996 1997							
Consumption	5	6	7					
Emissions	3	4	4					
Ems. in million t CO_2 equivalent 0.038 0.045 0.046								

The difference between consumption and emissions is due firstly to the only partial chemical conversion in the plasma reactor, and secondly to the effect of downstream flue gas purification systems.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

The assumptions for Scenarios I and II are set out in Part IV. Here only the resultant emissions are presented.

Table 28 presents the emissions that result in the year 2010 in business-as-usual Scenario I:

Table 28: 1995-2010 HFC-23 emissions from semiconductor manufacturingin business-as-usual Scenario I (forecast)								
	1995 2000 2005 2010							
Emissions [t HFC-23]	3	5	6	8				
Million t CO ₂ equivalent	0.04	0.05	0.07	0.10				

3 2010 Relative Emission Reductions - Scenario II

Table 29 shows the relatively lower emissions that result in Scenario II:

Table 29: 1995-2010 HFC-23 emissions from semiconductor manufacturing								
in reduction Scenario II (forecast)								
	1995 2000 2005 2010							
Emissions [t HFC-23]	3	4	4	1				
Million t CO ₂ equivalent	0.04	0.05	0.05	0.01				

In Scenario II, emissions in the year 2010 are reduced by 7 t. The global warming contribution is then only 0.01 million t instead of 0.10 million t CO_2 equivalent (Scenario I), which is a reduction by 86%.

8 Other HFC Applications

'Other HFCs' include the generation of HFC-23 as a by-product of the manufacture of HCFC-22. In both of the production plants in Germany, HFC-23 is captured and either processed further or - this being the greater part - thermally destroyed. Since 1996, HFC propellants in industrial aerosols are the most important in quantitative terms. Since 1998, HFCs are also in use for fire protection, and from 1999 onwards a limited use of HFCs is to be expected as solvents for precision cleaning.

1 1995-1997 Consumption and Emissions in Germany

Table 30: 1995-1997 consumption and emissions in Germany of 'other HFCs' in t/a HFC and million t CO ₂ equivalent								
	1995 1996 1997							
	Consump.	sump. Emission Consump. Emission Consump. Emiss						
HFC-23 (HCFC-22)	-	100*	-	0	-	0		
Industrial aerosols	100**	100	100	100	100	100		
Total	100	200	100	100	100	100		
Mill. t CO ₂ equiv.		1.288		0.118		0.118		

* Due to termination of emissions from mid-1995 onwards, only half of previous annual emission. ** On average, industrial aerosols contain 90% 134a and 10% 152a.

1.1 HFC-23 as By-Product

HFC-23 used to be released as a by-product of HCFC-22 manufacture. HCFC-22 is produced as a refrigerant and as a chemical feedstock for the plastic PTFE. According to figures reported by Hoechst AG in 1996, the HFC-23 released from production facilities to the atmosphere amounted to approx. 1-2% of HCFC-22 production. Since the CFC destruction plant (now operated by Solvay) commenced operation, surplus HFC-23 from the two HCFC-22 production facilities in Germany is consigned here to incineration to hydrofluoric acid (Solvay 1998). A small part is captured and marketed either as refrigerant R-23 or - after further distillation treatment - as etching gas for the semiconductor industry.

In 1995, HFC-23 by-product emissions amounted to half of the normal emissions of the previous years: 100 instead of 200 t. Due to the high GWP of HFC-23 (11,700), the global warming contribution in 1995 was significant: 1.17 million t CO_2 equivalent. Cf. Table 30.

1.2 Industrial Aerosols

75% of non-medical aerosols are used in personal care or in the household (IGA 1999). The aerosol branch, which in the 1980s was still the main consumer of CFCs, now uses as propellant almost exclusively propane, butane, dimethyl ether etc. Ozone-depleting substances are no longer used. HFCs, too, are scarcely used - apart from in

PU one component foam cans (cf. Section 3), they are only used in a small sub-sector classified as 'industrial sprays'. These are mainly freezer sprays and compressed-air sprays, which consume a total of approx. 100 t HFC annually (Meeh 1999). Cf. Table 30.

- Freezer sprays are used mainly for fault locating in electronic equipment. Through brief local cooling of live components, fault locations (e.g. in TV sets) can be identified more readily than when they are switched off (Electrolube 1997; Meeh 1998). As freezer sprays are used in live equipment, they must be nonflammable. Some 5-10% of freezer spray usage is in other areas, for instance in dental medicine for vitality testing of teeth or in public transport vehicles and cinemas to remove chewing gum from seats.
- In the applications of relevance here, 'compressed-air sprays' do not contain compressed air, but a compressed HFC used to blow dust away. These are used in electronics and optics where dust can cause malfunctions. Switchgear cabinets are a typical application. Because of potential ignition sources, HFCs are used here instead of compressed air due to their poor flammability.

Since some years, HFC consumption for industrial sprays has remained at a relatively constant level of 100 tonnes, consisting on average of 90% 134a and 10% 152a (estimate: Meeh 1999)¹⁶. Here the HFCs are not only propellants, but also process gases. The global warming contribution figures 0.120 million t CO_2 equivalent.

2 2010 Emissions Forecast

In departure from the previous sections, we do not generate two emission scenarios for 'other HFCs'. The main applications - industrial aerosols, solvents and fire protection - are impeded in their expansion, inter alia by statutory requirements, so that it makes little sense to distinguish between business-as-usual and relative emission reductions. HFC-23, which, as a by-product of chemical industry, had an important global warming contribution, is no longer emitted since 1996 and therefore not reducible, as long as we neglect - as everywhere else in this study - the small volumes that escape from recovery and filling facilities in all plants. Instead of two emission scenarios, we provide only one emissions forecast showing the most probable development until the year 2010.

The HFC applications already existing in 1997 were joined in Germany in 1998 by HFC-227 as a fire fighting agent. From 1999 onwards, the industrial aerosols include in addition to freezer sprays and compressed-air sprays also HFC-based cleaning sprays. The global warming contribution of 'other HFCs' in 1995, dominated by emissions of the by-product of HCFC manufacture, will remain far below that level for the foreseeable future.

¹⁶ The addition of 152a is necessary because 134a would on its own generate an impermissibly high interior container pressure of 12 bar at 50°C.

2.1 New: HFCs for Fire Protection

Halons, which were permitted for fire protection until 1991, have largely been replaced since then by environmentally friendly substances. Fixed systems for flooding indoor spaces now mainly use inert gases (nitrogen, argon) instead of halon 1301 (Minimax 1996; Total Walther 1996). Portable extinguishers for targeted fire fighting now contain powder, CO_2 or foam instead of halon 1211.

Of the halon replacement proposals originating in the USA (HFC-23 and 236fa, HFC-227, and the fluorocarbons 218 and 3110), HFC-227 was officially approved in Germany in 1997. This is produced by Great Lakes Chemicals in the USA and marketed by licensed companies under the trade name FM-200, in Germany by Kidde-Deugra (Stamp 1999). FM-200 extinguishes in the same way as halon 1301 through gas flooding of the room. For environmental reasons, Kidde-Deugra dispenses in installation and maintenance with discharge testing, minimizes false alarms through the installation of two detectors instead of one, and commands over a recycling facility for the extinguishing fluid. Over the medium term, a maximum FM-200 bank in Germany of 200 t is assumed, which could be reached by 2010 (Kidde-Deugra 1999).

Annual releases through actual fire fighting, false alarms and leakage are estimated at "less than 1%" (Kidde-Deugra 1999).¹⁷ It follows that of the HFC-227 sold since 1998, about 2 t will be released in the year 2010 from extinguishing systems (cf. Table 31), of this half in actual fires. The global warming contribution is 0.006 million t CO_2 equivalent.

Table 31: 1995-2010 'other HFC' emissions in t HFC and million t CO₂ equivalent							
Sub-sector HFC 1995 2000 2005 2010							
1. HCFC-22 by-product 23 100 0 0 0							
2. Ind. aerosols/solvents* 134a 100 130 200 200							
3. Fire protection 227 0 > 0 1 2							
Total		200	130	201	202		
Million t CO₂ equivalent 1.288 0.158 0.251 0.254							

* Prior to 2000: 90t 134a and 10 t 152a. After then: 120 t 134a, 70 t 43-10, 10 t 152a.

2.2 New: HFCs as Cleaning Solvent

New, nonflammable fluorinated compounds for precision cleaning and electronics cleaning also originate in the USA. The reason for this is that in the USA the solvents CFC-113, HCFC-141b and 1,1,1 trichloroethane are only banned since 1996 (banned in Germany already since 1.1.1993). The main products marketed are HFC-43-10mee = $C_5H_2F_{10}$ (Vertrel XF) and hydrofluoroethers (HFE-7100, HFE-7200).

Old CFC installations can continue to be used with HFEs as drop-in replacements. In Germany, however, HFEs basically come too late, as water-based cleaning agents and organic solvents based on alcohol and glycol ether have established themselves as replacements for CFC-113 and 1,1,1 trichloroethane. The market potential for HFEs is restricted to special applications and is not quantified here.

¹⁷ Cisneros/Robin (1999) also estimate approx. 1%.

In Germany, the use of HFC-43-10 in installations is banned by law. Under the 2nd Statutory Ordinance (2. BImSchV) pursuant to the German Emission Control Act (Bundes-Immissionsschutzgesetz), "no other volatile halogenated hydrocarbons" are permitted other than tetrachloroethene, trichloroethene and dichloromethane.¹⁸ Nonetheless, what is not permitted for installations (> 10 I charge) need not be prohibited in compressed gas containers, i.e. in aerosol cans.

2.3 Industrial Aerosols with Liquid HFCs

Fillers of industrial sprays not only expect the HFC market for freezer sprays and compressed-air sprays (cf. Section 1.2) to remain constant at a level of 100 t per annum. They further expect liquid HFCs to be used in the future in cleaning sprays on a scale of approx. 70 t per annum (Meeh 1999).

Dispensing with HFCs for on-site cleaning of live components in electrical engineering means that plants need to be switched off for cleaning if soiling causes a malfunction. In plants with high levels of soiling, this is a technical and economic problem. This is exemplified by the construction and mining industries and the metal processing industry, with high arisings of liquid mists or grinding dusts. The non-conductive organic solvents sometimes used are flammable and thus basically risky. Even if optimum results cannot be achieved with HFCs because of poor cleaning effect, plants can nonetheless be 'saved' across a shift and then thoroughly serviced during normal plant shutdown periods (Meeh 1999).

The market for fluorinated solvents for safe rapid cleaning without shutdown is estimated at approx. 70 t per annum. Due to their high price, HFEs scarcely come into question here, but HFC-43-10 does. A similarly nonflammable propellant is necessary in order to propel the nonflammable solvent from the aerosol can. This expands the market for HFC-134a by approx. 30 t per annum. At 1300, the GWP of 43-10mee is the same as that of HFC-134a. The additional global warming contribution of 70 t HFC-43-10 and 30 t HFC-134a amounts to 0.130 million t CO_2 equivalent.

It must be noted that in future, further applications may be developed for HFC-based solvents. If, for instance, the routine cleaning of medium-voltage switchgear of energy utilities should be carried out in future in live state - at present these plants are shut down - consumption would rise rapidly. This example serves as a warning that the emission forecasts presented in this study should not be misunderstood as upper limits.

3 Global Warming Contribution

The global warming contribution from 'other HFCs' in the year 2010 (anticipated to be 0.254 million t CO₂ equivalent) comes almost exclusively from industrial aerosols. In comparison, emissions from fire protection equipment are negligible.

¹⁸ It had been expected that the Ordinance would also exclude HFEs in installations. However, "hydrofluoroethers" are not subsumed under "volatile halogenated hydrocarbons" (source: German Federal Environment Ministry (BMU), reference number IG, I2-50121/29, 27.1.1998 - communicated by the company 3M Deutschland GmbH). HFEs do, however, fall as "halogenated organic solvents" under the emissions limitation of the European Union VOC solvent directive due soon to enter into force.

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II. Sulphur Hexafluoride (SF₆)

Overview

 SF_6 is the gas with the largest global warming potential, calculated by the IPCC in 1995 to be 23,900 CO₂ mass equivalent (based on a 100 year time horizon). Since the early 1990s, international climate research has been giving greater attention to this gas and its emission sources.

The greater part of SF_6 production worldwide is used in electrical equipment, notably switchgear. The present study on SF_6 usage in Germany confirms electrical equipment for power supply as the largest individual consumer, but comes to the conclusion that the structures of annual emissions are quite different from those of annual consumption. At present, most emissions come from car tires and, over the longer term, from soundproof windows.

In order to assess future SF_6 emissions from Germany, two scenarios extending until the year 2010 are elaborated in each application sector. Each of these scenarios is possible and realistic.

Scenario I is a business-as-usual scenario and extrapolates present trends. Scenario II is a relative reduction scenario, in which existing technology potentials for abating or substituting emissions are exploited in each individual SF_6 sub-sector. In Scenario II, only emission reduction measures are included whose ecological effect is not neutralized by ecological disadvantages elsewhere.

In Aggregate Scenario I - composed of six sectoral business-as-usual scenarios emissions drop from 1995 to 2010 from 260 t to 210 t (80%). In Aggregate Scenario II, emissions drop over the same period to 124 t (47%). In terms of global warming impact, Scenario I leads to about 5 million t CO_2 equivalent in the year 2010, Scenario II to 3 million t. This means a reduction by 2 million t CO_2 equivalent, or by 41%.

1 2010 Business-as-Usual Emissions Forecast - Scenario I

Scenario I is based for 1995-2010 upon the following sector-specific assumptions:¹⁹

- In <u>electrical equipment</u>, in both Scenarios I <u>and</u> II gas losses at manufacturers remain constant, while losses at operators rise by 0.2 t annually due to the growing stock in Germany. Decommissioning of annual cohorts only commences in 2010. SF₆ from old equipment is then recovered.
- In <u>soundproof windows</u> new annual production is constant at 2.6 million square metres of glazing. The SF₆:argon ratio in the filling mixture remains constant at 30:70%. From 2000 onwards, the first SF₆-filled windows installed in the 1970s are decommissioned. The SF₆ still in the glazing is not captured, but released.
- In <u>car tires</u>, whose gas filling is emitted completely after three years, the consumption level reached in 1998 (30 t) remains constant at least until 2007. Emissions thus remain at this level from 2001 to 2010.

¹⁹ The sectors 'electrical equipment' and 'other applications' are not examined in two scenarios. Their emissions are equal in both aggregate scenarios.

- 4. <u>Magnesium casting</u> output is expected to grow by 20% annually. However, owing to modern furnace technology, specific consumption of SF₆ as cover gas is relatively small in the newly established casting companies. A high specific SF₆ usage per t Mg only remains in old, small companies, which produce 1% of magnesium casting.
- 5. In <u>semiconductor manufacturing</u>, emissions grow in step with the 6% annual growth rate of consumption in Germany for wafer etching.
- 6. In <u>'other applications'</u>, emissions in both Scenarios I and II remain essentially stable in the eight sub-sectors, with the exception of the 'miscellaneous' sub-sector.

Under these assumptions, the emissions listed in Table 32 result for the period until 2010. Total emissions drop from 1995 to 2000 from 260 to 154 tonnes, but then rise again due to increased emissions from soundproof windows, reaching 210 t in the year 2010.

Table 32: 1995-2010 sectoral SF ₆ emissions in business-as-usual Scenario I									
in t/a									
1995 2000 2005 2010									
1. Electrical equipment	25	27	28	29					
2. Soundproof windows	108	60	90	126					
3. Car tires	110	50	30	30					
4. Magnesium casting	6	3	4	5					
5. Semiconductor man.	4	6	8	11					
6. Other applications	7	8	8	8					
Total	260	154	168	210					

- By far the most SF₆ emissions stem from <u>soundproof windows</u> (Table 32, line 2). In 1995, at 108 t, they are not quite as high as those from car tires, and in the year 2000 they reach a low of 60 t. After 2000, however, annual cohorts of glazing are due for disposal without capture of the gas filling. This is emitted completely to the atmosphere. In the year 2010, emissions from soundproof windows figure 126 t, and account for 60% of total SF₆ emissions in Germany.
- The marked drop of emissions from <u>car tires</u> (Table 32, line 3) from 110 t in 1995 to 30 t from 2001 onwards is the main reason why in the business-as-usual scenario total SF₆ emissions are lower in 2010 than in 1995. This drop in emissions is due to the circumstance that all the major tire retail chains have ceased to actively market SF₆ as a tire gas. Tire dealers only fill SF₆ instead of air or nitrogen upon the wish of their old customers.
- The emissions of SF₆-filled <u>electrical equipment</u> (Table 32, line 1) arise partly in their manufacture and partly in operation. The stock of this equipment in Germany is expected to rise until 2010. Emission will then total 29 t. If, as envisaged, a scheme is implemented to capture and reuse spent gas from decommissioned equipment in the subsequent years, a further rise in emissions is not to be expected from this application after the year 2010.
- As concerns the smaller applications magnesium casting, semiconductors and others emissions from magnesium casting are tending downwards owing to new

Table 33: 1995-2010 sectoral SF ₆ emissions in business-as-usual Scenario I									
in million t CO ₂ equivalent									
1995 2000 2005 2010									
1. Electrical equipment	0.60	0.65	0.68	0.70					
2. Soundproof windows	2.57	1.43	2.14	3.02					
3. Car tires	2.63	1.20	0.72	0.72					
4. Magnesium casting	0.15	0.07	0.08	0.11					
5. Semiconductor man.	0.10	0.15	0.20	0.27					
6. Other applications	0.18	0.19	0.19	0.19					
Total	6.22	3.69	4.01	5.01					

technologies (to 5 t in 2010), while 'semiconductors' rise to 11 t by 2010. 'Other applications' remain practically stable, growing 1995-2010 from 7 to 8 t.

Global warming emissions drop from 1995 to 2010 from 6.22 million t CO₂ equivalent to 5.01 million t, being 81% of the 1995 value (Table 33). Compared to the year 2000, in which emissions are substantially lower, this means a distinct rise from that year. In 2010, the bulk of emissions comes from soundproof windows, which are far in excess of emissions from car tires and electrical equipment.

2 2010 Emissions Reduction - Scenario II

The reduction scenario for 1995-2010 is based on the following sectoral assumptions:

- 1. While the annual new production of <u>soundproof windows</u> remains constant at 2.6 million square metres, new use of SF_6 as filling gas is phased out gradually by 2005, owing to changes in overall glazing design. This does not reduce the disposal losses that set in from 2000 onwards, but does reduce over the short term the filling losses and over the medium term the stock release losses.
- 2. By the year 2007, no SF₆ is used any more to fill <u>car tires</u>. The precondition to this is that the safety and environmental problems associated with this use are communicated to present customers.
- 3. Annual growth of <u>magnesium casting</u> output remains at 20%. Not only in the old casting companies but also in the newly established ones, SF_6 is replaced by SO_2 as a protective cover gas. SF_6 only remains in small-scale specialty applications.
- 4. In <u>semiconductor manufacturing</u>, from 2000 onwards new etching chambers are only installed with high-efficiency exhaust gas purification systems. From 2009 onwards, the old facilities, too, are replaced by new ones with devices for SF₆ destruction in the waste gas.
- 5. For <u>electrical equipment</u> and <u>other applications</u>, the assumptions and consequently the emissions remain the same as in Scenario I.

Under these conditions, the emissions shown in Table 34 result for the period until 2010.

- Compared to Scenario I, total SF_6 emissions sink not only to 210 t, but to 124 t. This is 86 t or 41% less.
- Developments in soundproof windows and car tires are crucial to emissions reduction in Scenario II. In both applications, new SF₆ use is phased out gradually

from 1999 onwards. No emissions occur any more from car tires from 2010 onwards. No filling losses arise from soundproof windows from 2006 onwards, so that in 2010 the emissions in this sector - consisting of disposal and stock release losses - no longer figure 126 t but only 84 t (Table 34, line 2).

Table 34: 1995-2010 sectoral SF ₆ emissions in reduction Scenario II								
in t/a								
1995 2000 2005 2010								
1. Electrical equipment	25	27	28	29				
2. Soundproof windows	108	54	51	84				
3. Car tires	110	50	17	0				
4. Magnesium casting	6	3	2	0.3				
5. Semiconductor man.	4	6	6	2				
6. Other applications	7	8	8	8				
Total	260	148	111	124				

 In magnesium casting, too, SF₆ is almost completely phased out due to conversion to SO₂. SF₆ emissions in 2010 approach zero, at 0.3 t.

- Despite 6% annual growth of SF₆ usage in semiconductor manufacturing, emissions in 2010 are below the initial level of 1995, owing to complete fitting of all new facilities with exhaust gas purification systems and the replacement of old facilities, the latter setting in from 2009 onwards.
- Emissions from electrical equipment and other applications are the same as in Scenario I (as assumed). Due to the phase-out of SF₆ in tire filling, electrical equipment is the second largest emission source in 2010, but still far behind soundproof windows.
- Soundproof windows account in 2010 for an even larger proportion of total SF₆ emissions - 68% - than in Scenario I, although reduced in absolute terms.
- Table 35 shows the development in terms of CO₂ equivalent emissions. In 2010, 2 out of the total of just under 3 million t are attributable to soundproof windows alone.

Table 35: 1995-2010 sectoral SF ₆ emissions in reduction Scenario II								
in million t CO ₂ equivalent								
1995 2000 2005 2010								
1. Electrical equipment	0.60	0.65	0.68	0.70				
2. Soundproof windows	2.57	1.28	1.21	2.01				
3. Car tires	2.63	1.20	0.40	0.00				
4. Magnesium casting	0.15	0.07	0.04	0.01				
5. Semiconductor man.	0.10	0.14	0.14	0.05				
6. Other applications	0.18	0.19	0.19	0.19				
Total	6.22	3.53	2.66	2.95				

The different aggregate evolution of emissions in Scenarios I and II over the entire 1995-2010 period is visualized in Diagram 22:



Diagram 22: 1995-2010 SF₆ emissions in Germany in Scenarios I and II in tonnes. After a marked drop until 2001, emissions in Scenario II remain stable despite the commencing disposal losses from soundproof windows from 2001 onwards. This establishes a growing reduction potential compared to Scenario I, in which emissions rise again from 2001 onwards.

Diagram 23 shows that in 2010, CO_2 equivalent emissions figure 5 million t in Scenario I, and 40% less in Scenario II, namely ca. 3 million t.



Diagram 23: 1995-2010 SF₆ emissions in Scenarios I and II in million t CO_2 equivalent. In Scenario I, emissions in 2010 figure more than 5 million t, in Scenario II only 3 million t.

Annex: The years 1995 to 1997

Tables 36 and 37 serve to complete the environmentally relevant emission data for the years 1996 and 1997.

1995-1997 emissions in t per annum

Table 36: 1995-1997 sectoral SF ₆ emissions in t/a						
1995 1996 1997						
1. Electrical equipment	25	28	27			
2. Soundproof windows	108	85	75			
3. Car tires	110	115	120			
4. Magnesium casting	6	3	3			
5. Semiconductor man.	4	5	5			
6. Other applications	7	8	8			
Total	260	243	238			

1995-1997 Global warming contribution

Table 37: 1995-1997 sectoral SF ₆ emissions in million t CO₂ equivalent						
	1995	1997				
1. Electrical equipment	0.60	0.66	0.64			
2. Soundproof windows	2.57	2.02	1.80			
3. Car tires	2.63	2.75	2.87			
4. Magnesium casting	0.15	0.07	0.07			
5. Semiconductor man.	0.10	0.11	0.13			
6. Other applications	0.18	0.18	0.19			
Total	6.22	5.81	5.70			

1 Electrical equipment

Electrical equipment for electric power supply is by far the largest individual SF_6 consumption sector in Germany, with just under 400 t per annum. However, due to the high export share of more than 80%, only 60-70 t SF_6 join the stock in Germany in new equipment and facilities. SF_6 emissions in Germany from the manufacturers and operators of electrical equipment figure approx. 25 t per annum. In the 1995-1997 period, electrical equipment was the third largest SF_6 emissions source within Germany.

1 SF₆-Filled Electrical Equipment: Switchgear and Transformers

 SF_6 is used above all in high-voltage (110-380 kV) and medium-voltage (10-30 kV) switchgear and controlgear. Here the gas functions both as an arc-quenching and insulating medium instead of air or mineral oil. It is not commonly used in the low-voltage (< 1 kV) range.

Switchgear, controlgear and the instrument transformers usually associated with them are located at the nodal points of electric supply networks, where energy is distributed from higher-level transmission lines to outgoing lines. In addition to switching operating current in sub-networks (often because of maintenance and repair work), switchgear needs to be capable of switching off within milliseconds the high short-circuit currents that result from faults and malfunctions in the circuit and quenching the high-ampere arcs that result at the switch contacts.

1.1 High- and Medium-Voltage Switchgear

The two most important high-voltage product groups containing SF₆, namely metalenclosed gas-insulated switchgear (GIS) and outdoor circuit breakers, are in use in Germany since 1965. SF₆ technology became dominant in the high-voltage sector between 1970 and 1974. In medium-voltage switchgear, SF₆ insulation was taken up later. The first SF₆-insulated load-break switchgear came on the market in 1978, the first circuit breakers in 1982.

1.2 High-Voltage Instrument Transformers

 SF_6 -insulated outdoor instrument transformers were introduced in 1978. For long, oilimpregnated paper was the dominant form of insulation of this type of equipment. In 1990, the leading German producer converted entirely to SF_6 , and at the other producer the relation between oil and SF_6 insulation shifted clearly in favour of the latter.

1.3 SF₆ Stocks in Germany in Electrical Equipment

The main operators of SF₆-containing electrical equipment, just under 90% of which are public electric utilities, had in 1995 a stock of 1000 t of this gas. "Of this, 89% are in high-voltage equipment and systems, whereby conventional equipment (circuit breakers, transformers) account for a third of the SF₆ stock, and gas-insulated switchgear (GIS) for two thirds. Only about 11% of the SF₆ stock in electric energy supply equipment is in medium-voltage systems." (VDEW 1996, 2-3)

In contrast to the stock in Germany, sales of SF_6 -filled switchgear in Germany are currently dominated by the medium-voltage sector (two thirds in 1996). The operators of these plants and equipment are not so much supra-regional electric utilities, but mainly large-scale industry and municipally-owned utilities (ABB 1999). In the early 1990s air was still mainly used as insulating medium in medium-voltage equipment.

2 1995-1997 Consumption in Germany

The annual SF₆ consumption of manufacturers for filling new electrical equipment (switchgear, transformers) rose over the 1995-1997 period by 40 t from 349 t to 388 t. Of this, switchgear manufacturers used 308-348 t and transformer manufacturers 40 t annually (cf. Table 38). Most of the gas was exported abroad together with the products.

Table 38: 1995-1997 SF₀ consumption in Germany in the manufacture of electrical equipment [t/a]							
1995 1996 1997							
Switchgear	308	345	348				
Instrument transformers	40	40	40				
Power transformers 1 <1 -*							
Total:	Total: 349 386 388						

Sources: ZVEI 1996 and UBA 1999; Transformer Survey 1999.

* Since the end of 1996, SF₆-filled power transformers are no longer produced.

3 1995-1997 Emissions in Germany

Emissions within Germany must be distinguished according to gas losses at the manufacturer and at the operator. Manufacturer emissions are plant emissions and assembly losses, operator emissions are leakage and maintenance losses.

3.1 Manufacturer Plant Emissions

In the manufacturing plant, losses arise in development, testing and manufacture of components or systems for sale in Germany and for exports. According to UBA 1999, switchgear plant losses figured 4.4% of total SF_6 consumption (see Table 38) in 1996 and 3.9% in 1997.

In order to restrict losses during filling of equipment and systems, SF_6 is not filled into the gas spaces through air displacement, by which it could partially escape with the exiting air; state-of-the-art is rather to evacuate the gas spaces prior to filling. This is motivated by the technical necessity of keeping the SF_6 gas free of air and moisture in order to thus minimize the generation of decomposition products by secondary reactions.

For transformers, manufacturers estimate plant losses caused by faulty equipment and other sources at only 2% of annual fillings (Transformer Survey 1999). One explanation given for this is that transformer manufacturers undertake only little own research and development effort of their own.

Table 39: 1995-1997 SF ₆ emissions in Germany from manufacturers and operators of electrical equipment [t/a]						
1995 1996 1997						
Manufacturer plant emissions	14.00	15.99	14.27			
Manufacturer assembly emissions	1.30	1.37	1.80			
Continuous operator losses*	9.90	10.35	10.90			
Total:	21390	2193	2219			
Ems. in million t CO ₂ equivalent	0.602	0.662	0.645			

Total plant emissions figured 16 t in 1996 and 14.3 t in 1997 (Table 39).

Sources for 1996 and 1997: UBA 1999; for 1995: own estimates.

* Leakage and maintenance losses (annual refills).

3.2 Manufacturer Assembly Emissions

Gas losses at the site of assembly are a further - smaller - source of emissions. They arise during assembly and commissioning of systems and equipment prior to hand-over to the operator. Plants and equipment are shipped to the site of assembly with either SF_6 filling at normal pressure or with nitrogen filling.

For operation at high voltage, pressure in the gas spaces must be raised to 3-4 bar, because when pressurized the insulating effect is about ten times better than that of air. It is this that makes it possible to keep distances between live components and thus overall dimensions up to ten times smaller. Space savings are one of the main arguments for using SF₆ technology. In the circuit-breaker compartment of gas-insulated switchgear (GIS), the operating pressure is even raised to 6.5 bar. This is the pressure at which SF₆ is just still in gaseous state at minus 25°C (ABB 1996).

Assembly losses during the filling of switchgear at the site of usage in Germany figured 2% to 2.6%. Transformer manufacturers state for their transformers erected in Germany (outdoor transformers) assembly losses of only 0.2% of the new filling.

Total assembly losses figured 1.37 t in 1996 and 1.80 t in 1997 (Table 39).

3.3 Leakages at Operators

Although the filled spaces of most metal-enclosed high-voltage SF_6 equipment are under pressures of more than 6 bar, gas losses due to leakage during operation are relatively low. A DIN Standard (DIN IEC 298 VDE 0670, p.43) permits annual leakage rates of 1%-3%, but in practice only older equipment manufactured before 1980 is in this range. Modern equipment is tighter, losing less than 0.5% annually, which must be refilled. All manufacturers guarantee for new equipment leakage rates of less than 1%.

3.4 Maintenance Losses at Operators

Maintenance losses arise during the routine or malfunction-related opening of equipment. Maintenance normally involves cleaning the gas of the moisture, air and chemical decomposition products that have collected, and checking and replacing

metallic contacts and components, and seals. Maintenance is carried out by the operator or by the manufacturer.

 SF_6 switchgear does not need much servicing, as its interior is protected against soiling and weather impacts (ice, snow, saline air). Moreover, discharge-intensive switching is not frequent, and high-ampere short-circuit arcs, for which the circuit breakers are designed, do not occur more often than every 1-2 years. Circuit breakers are designed to switch up to 10,000 rated currents and to switch off 50 to 100 short-circuits during their lifetime (VDEW 1994, 12). Thus a typical service plan for SF_6 outdoor circuit breakers envisages a first inspection opening of the equipment after 20 years of operation (Siemens 1996b; Siemens no year). Maintenance intervals for metalenclosed SF_6 equipment are similarly long.

Today, techniques are available by which to limit emissions arising when the gas space is opened during maintenance and repairs. On-site gas recycling is state of the art. Discharge and refill are in a closed cycle (VDEW 1993, 48, 49). For this procedure, operators and manufacturers have special mobile service units with which the SF_6 is suctioned off to an absolute pressure of 50 mbar, cleaned of gaseous and dust contamination, liquefied, consigned to interim storage and subsequently refilled (Neumann 1992, 269).

Annual losses in Germany at equipment operators due to leakage and maintenance figure 0.9% in real terms in relation to the total SF_6 stock contained in all equipment.²⁰ Operators include, in addition to public electricity suppliers, also industrial companies and the national railway operator. These have approx. 10% of the total SF_6 stock in electrical equipment in Germany.

Electricity suppliers and other operators

In Germany, electric utilities, industry and railway operators generated together in 1997 508 (1996: 511) billion kilowatt-hours electricity. Just under 89% of the electricity came from power plants operated by public utilities. Industrial power plants accounted for some 10% of total German electricity generation in 1997. Deutsche Bahn, the national railway operator, had a share of a good one percent with seven billion kWh (VDEW 1998b). Experts estimate that industry and the railway operator together have 10% of the total SF₆ stock in Germany in SF₆-filled equipment and systems (ABB 1996).

4 2010 Emissions Forecast

In departure from the other emission sectors, we do not elaborate two emission scenarios for SF_6 emissions from electrical equipment. A distinction between business-as-usual and reduction scenarios is not expedient for the period until 2010, as it is entirely improbable that emissions develop other than the emerging trend. This is because due to the 40-year lifetime of SF_6 -filled equipment, first annual cohorts of the

²⁰ VDEW, the German association of electric utilities, already determined an operator emissions rate of 0.9% in 1995. For the public electricity suppliers (excluding industry and the national railway operator) it declared: "Due to design-related leakage, gas losses in maintenance and malfunction-related losses, some 9000 kg SF₆ gas needs to be refilled annually in the electrical equipment employed in electric energy supply in Germany. In relation to the total quantity in use (1000 t; the authors) this corresponds to an average annual refill rate of 0.9%." (VDEW 1996)

first generation will only be due for scrapping after the year 2010. Then, however, the question will indeed arise of what happens with the SF_6 fillings - will they be recovered or discharged to the atmosphere? As the temporal scope of the present study ends at the year 2010, instead of two scenarios we provide here only a single forecast of the most probable development of emissions until that year. We then discuss conditions that guarantee SF_6 recovery from old equipment after 2010.

4.1 Future Plant Losses

For the foreseeable future, the switchgear and transformer industry does not assume any substantial increase in its production, so that over the medium term the total consumption of SF_6 can be assumed to remain in the region of 350 to 400 tonnes per year. It is thus not to be expected that plant losses - which are proportional to total SF_6 consumption - rise. They can be expected to be in the region of 14 to 16 t per annum (centre: 15 t).

4.2 Future Site Assembly Losses

The site assembly losses of manufacturers are not a function of total annual SF_6 consumption, but depend only upon the proportion of this which is destined for use within Germany. Medium-term growth rates of electricity consumption in Germany are estimated at "not more than one percent per year on average" (VDEW 1998a). New equipment in the high-voltage network of the supra-regional electricity suppliers serves primarily to replace retired air- or oil-insulated equipment built prior to 1970-1980 by SF_6 -filled equipment. The equipment stock will scarcely grow.

The slow-down in inland sales of high-voltage equipment is compensated by mediumvoltage equipment for industry and municipally-owned utilities. As operating stresses are lower compared to high-voltage equipment, SF_6 -filled medium voltage equipment is not or only slightly pressurized. This is one reason why their filling in the plant and their site assembly at the operator are viewed as less emissive than that of high-voltage equipment (ABB 1999).

No rise in site assembly losses is expected over the medium term. Emissions are expected to be in the range of 1.3 to 1.8 t per annum (centre: 1.5 t).

4.3 Future Operator Emissions

Operator losses are the only emission category that will grow slightly until 2010: from 9.9 t in 1995 to approx. 12.8 t.

 SF_6 losses at the operator depend upon the age structure and the size of the equipment stock.

A VDEW study carried out in the year 1996 gives an insight into the impacts of the age structure of the equipment stock upon leakage losses. In 1995, the refill quantities for SF₆-insulated switchgear built in 1980 and earlier accounted for almost half of the entire refill quantity, although these old units only make up less than 20% of the total SF₆-filled stock. "This corresponds to an annual refill rate of 2.2%. Refill quantities for SF₆-insulated switchgear built after 1980 were substantially lower. These components

achieved annual refill rates of 0.3% to 0.6%." (VDEW 1996) Equipment of the second and third generation has even better tightness. As concerns the medium-voltage equipment, with its growing sales prospects since 1996, annual operating emissions are calculated to be even lower than those of modern high-voltage equipment (ABB 1999).

If present usage trends continue then the total stock in Germany will grow between 1996 and 2010 by approx. 50 t net per annum. This leads until 2010 to 1850 t, being 750 t more than the 1100 t stock of 1995.

The composition of stocks according to vintage is decisive for operation emissions:

- For the new equipment that swells the German stock by 750 t over the 1996-2010 period, an annual operator emissions rate (leakage plus maintenance losses) of 0.4% is assumed.
- According to VDEW 1996, public electricity suppliers, industry and the railway operator have a stock of equipment built in the 1980-1995 period with a total filling of 900 t. For this, an annual emission rate of 0.6% is assumed.
- Equipment built before 1980 has a stock of 200 t, and an annual emission rate of 2.2%.

From this, the growing operator stocks and emissions shown in Table 40 follow for the years 2000, 2005 and 2010:

Table 40: 1995-2010 SF ₆ emissions from operators of electrical equipment							
in t/a,	accordin	g to equip	ment age	e (forecast	:)		
	2000 2005 2010						
Year of manufacture	Stock	Emission	Stock	Emission	Stock	Emission	
Prior to 1980 ER: 2.2%	200 t	4.4 t	200 t	4.4 t	200 t	4.4 t	
1980-95 ER: 0.6%	900 t	5.4 t	900 t	5.4 t	900 t	5.4 t	
1996 ff. ER: 0.4%	250 t	1.0 t	500 t	2.0 t	750 t	3.0 t	
Total	1350 t	10.8 t	1600 t	11.8 t	1850 t	12.8 t	

ER = annual emission rate of the stock

4.4 Total of Manufacturer and Operator Emissions Until 2010

Table 41 and Diagram 24 show that from 1995 to 2010 the annual total of manufacturer and operator emissions in Germany can be expected to rise by approx. 3 t - to just under 30 t (calculated value: 29.3 t). Manufacturer emissions in the plant and during site assembly remain stable, figuring throughout the 2000-2010 period 15 and 1.5 t respectively. It remains to be noted that due to the high export share of German switchgear manufacturers, the majority of plant losses is attributable to equipment produced for export.



Diagram 24: 1995-2010 annual SF_6 emissions from electrical equipment in t (forecast). While manufacturer emissions (15 t plant and 1.5 t site assembly losses) remain constant, operator emissions grow slightly. The low rate of growth is due to the circumstance that newly installed equipment has only low annual emission rates (0.4%). Replacement of the more emissive old equipment (2.2% emission rate) commences after 2010. Only then do continuous operator emissions drop over a 10-year period. However, decommissioning emissions then occur - these are not shown in the diagram, as they are outside of its temporal scope.

The growth in total emissions is driven by operator emissions. These are not yet decommissioning emissions, but gas losses through leakage and maintenance during continuous operation. Because the electrical equipment newly installed from 1996 onwards is less emissive than older equipment, the absolute volume of emission growth is relatively low.

Table 41: 1995-2010 total annual SF₅ emissions from electricity supply equipment (forecast)						
	1995	2000	2005	2010		
Plant losses [t]	14.0	15.0	15.0	15.0		
Site assembly losses [t]	1.3	1.5	1.5	1.5		
Operator losses [t]	9.9	10.8	11.8	12.8		
Total [t]	25.2	27.3	28.3	29.3		
Million t CO ₂ equivalent	0.602	0.652	0.676	0.700		

Table 41 shows that total SF_6 emissions from the manufacture and operation of electrical equipment rise from 25.2 t in the year 1995 to 29.3 t in the year 2010.

The global warming contribution of these emissions rises over that period from 0.6 to 0.7 million t CO_2 equivalent (cf. Table 41).

5 On the Prevention of Decommissioning Emissions After 2010

Due to the long lifetimes of regularly serviced equipment, the first annual cohorts of the first equipment generation will be decommissioned only after 2010. Until then, decommissioning and the associated emissions will only occur in special cases, such as when whole units and plants are destroyed by external impacts. When SF_6 -filled equipment begins to be retired on a large scale from 2010 onwards, it is essential that the gas is suctioned off prior to scrapping. It can then be regenerated on site or at the equipment manufacturer for refilling, using normal service equipment. Where it is excessively soiled, reprocessing at the gas producer is possible.

Electrical equipment is as yet the only SF_6 application for which a concept for the reuse of spent SF_6 gas has been developed. This was developed by Solvay Fluor und Derivate GmbH, an SF_6 manufacturer, in co-operation with DILO Armaturen und Anlagen GmbH, an SF_6 maintenance and measuring equipment company. In addition to on-site recycling by mobile maintenance units, it provides for the reprocessing of spent gas by the SF_6 manufacturer (e.g. Solvay), who "regenerates spent gas to new gas that corresponds in all points to the DIN IEC 376 standard for new gas" (Solvay 1996). SF_6 which cannot be reused is consigned to disposal.

The German operator and manufacturer associations - VDEW and ZVEI - committed themselves in 1996 to this type of emission limitation in a joint declaration on the use of SF_6 in electrical switchgear in Germany (VDEW/ZVEI 1996).

The aim of the reuse concept is to guarantee that in the course of equipment decommissioning, which will commence on a larger scale from 2010 onwards, the annual SF_6 fillings of more than 50 t are captured and consigned to reprocessing or disposal with minimum recovery losses.

2 Soundproof Windows

Soundproof windows are the second largest consumption sector of SF₆ after electrical equipment, and are - over the long term, too - by far the largest emission sector.

1 SF₆ in Soundproof Windows

In the 1970s, single glazing in windows and glass facades was superseded by soundinsulating double glazing, into which SF_6 has been filled since 1975 in order to improve the sound-insulating effect in the pane interspace. Compared to glazing filled with air or noble gases, SF_6 reduces the sound transmission intensity in the frequency range of relevance to building acoustics by 2-4 dB (Koch 1998). Approx. 5 dB is perceived by people as a halving of noise (Schumacher 1999).

The sound-insulating effect results from the reduced speed of sound, which is only 130 m/s in SF₆ compared to 330 m/s in air (Holler 1995, 82). In a first approximation, the speed of sound sinks in proportion to the square root of the molecular weight of a gas (Derner 1992, 60). SF₆, being a very heavy gas, is therefore suited for sound insulation - but not for thermal insulation. In the latter function, SF₆ is poorer than air (Huntebrinker 1995, 43). Argon is the gas used for thermal insulation in glazing.

The function and lifetime of all insulating glazing depends decisively upon the gastightness of the edge seal design. The outer sealing of the glazing rim must prevent both outward gas losses and the inward penetration of water vapour for as long as possible. This is done by pressing the rim of the panes along the whole length against an adhesive-coated metal bar (the spacer) and sealing this all around with sealing mass. The adhesive used is polyisobutylene, the sealant is polysulphide.



Fig. 2: Edge seal of double-glazed soundproof window with SF_6 -filling for improved sound insulation. The metal spacer around the rim of the panes is sealed with a polysulphide mass against inward and outward diffusion. The desiccant binds air moisture that has diffused inwards for 20 to 30 years (drawing after Holler 1995, 69).

In order to balance internal pressures, the spacer has a slit permitting diffusion and contains a desiccant to bind any water vapour that has entered into the interspace between the panes (see Fig. 2).

The durability of soundproof windows depends primarily upon the organic adhesive and sealant, specifically upon the stresses imposed upon these by weathering and UV/IR irradiation with subsequent cracking, and imposed by penetrating water vapour from moist window frame material (PVC, aluminium, wood) in which the sealed glazing is set. Depending upon geography and climate, the normal lifetime of soundproof windows is between 15 and 30 years (Soundproof Window Survey). The most important limiting factor is the diffusion of water vapour, which gradually exceeds the capacity of the desiccant to bind it and to maintain a moisture balance in the interspace, with the result of inside condensation at lower temperatures (Holler 1995, 92).

2 1995-1998 Consumption and Emissions in Germany

Because of the close supply connection to the window and glass facade construction branch, the production of soundproof windows, using industrially manufactured basic glass, is decentral and in smaller enterprises. The number of producers in Germany is estimated at 300 to 400. External trade is marginal (Soundproof Window Survey).

2.1 1995-1998 SF₆ Consumption

On average over the 1995-1998 period, approx. 36 million square metres of insulating glazing were fabricated annually in Germany, of which approx. 2.6 million square metres was SF_6 -filled soundproof glazing. Since 1975, the year of the first SF_6 fillings, the average pane interspace has grown from 12 to 16 millimetres. This results for the 1995-1998 period in a gas filling volume of soundproof windows amounting to some 42 million litres.

Two points need to be taken into consideration when determining the amount of SF_6 required for this:

- <u>Firstly</u>, the filling ratio of the interspace with gas only averages 90%, as 10% residual air can scarcely be eliminated during the filling process.
- <u>Secondly</u>, the share of SF₆ in the filled gas is dropping. Until the mid-1980s, soundproof windows were filled exclusively with SF₆. Since then, thermal insulation requirements are rising (3. Wärmeschutzverordnung 1994). This is why many soundproof windows are increasingly receiving blends of argon and SF₆. Over the 1995-1998 period, the average proportion of SF₆ to argon in soundproof windows dropped from 75% in 1995 to 33% in 1998. It may be assumed that in 1999 a gas composition of 30% SF₆/70% argon will have established itself as a longer-term compromise between acoustic and thermal insulation.

The drop in the specific SF_6 share in gas filling from 75% to 33% over the 1995-1998 period had a strong impact on annual overall consumption (the quantity sold by the gas trade), as shown in Table 42. Annual consumption dropped over that period from 277 to 121 t - by 56%.

1005 1006 100	
1995 1996 1997	' 1998
SF ₆ consumption [t] 277 205 169	121

Source: Gas Trade Survey 1999.

2.2 1995-1998 SF₆ Emissions

 SF_6 emissions from soundproof windows must be distinguished according to three different categories, whose quantitative impact changes over time. <u>Filling losses</u> occur exclusively in the year of fabrication and are directly proportional to annual SF_6 consumption. <u>Stock leakage emissions</u> are the gas losses from the filled glazing throughout its entire lifetime, which averages 25 years. The emissions of the third category, <u>disposal losses</u>, only occur at the end of the use phase of the glazing - 25 years after the filling emissions.

2.2.1 Filling Losses (Overfilling)

Of the annual SF_6 consumption for soundproof windows, 33% arise as filling emissions in the year of fabrication. Both in hand-held equipment and in automatic gas filling presses, turbulence in the interior is unavoidable, so that not only residual air but also an air- SF_6 mixture escapes, and all the more the further the filling process progresses. The resultant gas loss, the overfilling, ranges from 30 to 60% of the fill. A survey of leading producers of sound-insulating glass and of glass filling equipment reveals that on average the overfilling amounts to 50% of the effectively filled quantity (Soundproof Window Survey). As noted above, the latter only amounts to 90% of the interspace.

2.2.2 Stock Leakage Emissions

Stock leakage emissions are assumed in this study to be 1% per annum. This contains both continuous gas losses through the edge seal and gas losses through glass breakage before and during use. A DIN standard prescribes for gas losses during the use phase a maximum limit of 1-1.2% (DIN 1286, T 2). All surveyed experts stated unanimously that the real SF6 gas loss in intact glazing is below this permissible limit (Soundproof Window Survey). However, further gas losses through glass breakage do occur, which can happen during transport, installation and the 20-30-year use phase.

2.2.3 Disposal Losses

While a reprocessing concept for disposal of filled equipment is currently being introduced for SF₆ in electrical equipment, in the soundproof window sub-sector recycling schemes are only in place for the frame materials PVC and aluminium, and partially for the glass (Koos 1996; Brüggemann 1996). There are no plans for a scheme to recover SF₆ from end-of-life glazing, and this is not considered technically feasible (Koch 1999; Schumacher 1999). It thus must be expected that at the end of the lifetime of the soundproof glazing or of the window frame the filling gas escapes to the atmosphere. These are major emissions, as of the initial SF₆ filling only the stock leakage losses of 1% per annum over the 25-year lifetime are absent, and thus approx. 78% are still present. As the first SF_6 soundproof windows were installed in 1975, the first annual cohorts of disposal and resultant gas losses are to be expected from the year 2000 onwards. These will then increase from year to year, peaking in 2020 at 142 t.

2.2.4 1995-1998 Quantity and Composition of Emissions

Table 43 shows that over the 1995-1998 period total emissions dropped from 108 to 60 t. This drop was exclusively an outcome of sinking filling losses (line 1), which developed proportionately to the - declining - consumption in each year. Stock leakage emissions (line 2) grow steadily but slowly. Disposal losses (line 3) only commence in the year 2000.

Table 43: 1995-1998 SF ₆ emissions from soundproof windows in t/a SF ₆							
and in million t CO ₂ equivalent							
1995 1996 1997 1998							
1. Filling losses*	92	67	57	40			
2. Stock leakage losses**	16	18	19	20			
3. Disposal losses ***	-	-	-	-			
Total emissions	108	85	75	60			
Million t CO ₂ equivalent	2.57	2.02	1.80	1.43			

* = 33% of the consumption of the current year. ** = 1% of the stock accumulated since 1975. *** Disposal losses only occur from 2000 onwards.

Over the 1995-1998 period, the global warming contribution of total emissions dropped from 2.57 to 1.43 million t CO_2 equivalent.

The drop in emissions over the 1995-1998 period is only a transient phenomenon. Disposal losses will lead to total emissions rising again in the future, even if no new SF_6 were used at all with immediate effect.

3 2010 Business-as-Usual Emissions Forecast - Scenario I

In Scenario I it is assumed that by the year 2010, new annual production of soundproof glazing figures 2.6 millions square metres or 6-7% of all insulating glazing. Moreover, the following assumptions are made:

- 1. In combined thermal and acoustic insulation glazing, the gas ratio of SF₆ to argon figures 30 to 70 from 1999, and remains so over the medium term. This results in constancy of annual consumption and annual filling emissions until 2010.
- 2. Data are available on annual SF₆ consumption since 1975. These data and the SF₆ consumption levels forecast until 2010 result in the accumulated SF₆ stocks, from which 1% SF₆ escapes annually.
- 3. With a delay of 25 years, being the lifetime of the glazing, emissions occur in disposal which are not captured. In the year 2000, the production year 1975 is disposed of, in the year 2001 the production year 1976 etc. Here it needs to be considered that the initial filling is reduced by the annual leakage rate of 1%.



Diagram 25: 1995-2010 SF₆ emissions from soundproof windows in Scenario I [t/a]. After the drop in total emissions over the 1995-1999 period down to 57 t due to declining filling emissions (black area) total emissions rise again from 2000 onwards due to disposal losses (grey area). In 2008, the historic peak of 1995 is exceeded. In 2010, emissions figure 126 t (67 t disposal loss + 23 t stock leakage loss + 36 t overfilling).

Under these conditions, the emissions shown in Diagram 25 and Table 44 result for the 1995-2010 period:

- From 1995 to 1999, total emissions drop to 57 t due to the decline proportional to declining new consumption of filling emissions.
- Disposal losses arise for the first time in 2000. These then become the largest emissions item, growing to 67 t by 2010. As assumed, filling emissions remain constant from 1999 onwards, namely at 36 t. Stock leakage emissions remain relatively stable at 22-23 t.
- In Scenario I, total emissions in the year 2010 figure approx. 126 t.²¹

Table 44: 1995-2010 SF ₆ emissions from soundproof windows in							
business-as-usual Scenario I [t/a]							
1995 2000 2005 2010							
1. Filling losses	92	37	37	37			
2. Stock leakage losses	16	20	21	23			
3. Disposal losses	-	3	31	66			
Total emissions	Total emissions 108 60 89 126						

²¹ The peak in the year 2010 figures 197 t total emissions, of which 142 t are from disposal.

4 2010 Relative Emission Reductions - Scenario II

In view of the large SF_6 stock in already installed soundproof windows (1998: 2000 t), a substantial reduction in emissions over the medium term is impossible if gas recovery at disposal is not implemented. Such recovery appears unrealistic today. Nonetheless, by phasing out SF_6 in new windows, the considerable filling emissions could cease over the short term and stock leakage emissions would also drop.

Sound reduction is not brought about primarily by SF_6 , but by overall glazing design. The factors here are increased interspace, different glass thicknesses, or the use of cast-resin compound glass. This achieves sound reduction factors of 35 to 50 dB. SF_6 can enhance sound reduction by an additional 2-4 dB, but is not a substitute for advanced glazing design (Koch 1999; Schumacher 1999).

When evaluating SF_6 , it is essential to distinguish between sound reduction measured for the glazing on its own and that for the whole installed window unit.

According to a new research project of the Institut für Fenstertechnik (i.f.t.) institute for window technology in Rosenheim "the weighted sound reduction factor of glazing with partial SF_6 filling is approx. 4 dB higher than in argon-filled glazing of the same design." However, this no longer holds for the whole window: "In windows with SF_6 -containing glazing, the weighted sound reduction factor is improved by approx. 1 dB. ...We also found designs in which the sound reduction factor was reduced by 1 dB." (i.f.t. 1999).

The i.f.t. institute follows from this: "These findings render the use of SF₆ debatable, suggesting the withdrawal of SF₆ filling gas from the market" (loc. cit.). For: "The improved sound reduction in soundproof windows offered by SF₆ or SF₆ blends can be compensated for by increasing glass thickness or by increasing the pane interspace by approx. 2-4 mm".²²

 SF_6 improves the sound reduction effects of glass thickness, glass type and pane interspace in the glazing on its own by at most 4 dB, but in the whole installed window by only 1 dB. A further aspect is the limited frequency range in which SF_6 has a reducing effect. The Fraunhofer Institute for Building Physics notes that the weighted sound reduction factor pursuant to DIN 52210 of August 1984 weights low frequencies, which dominate in urban traffic noise, less than in the more recent European standard DIN EN ISO 717, Part 1 (Koch 1998). SF_6 has a "distinct drop in effectiveness at low frequencies ". With the modified European standard, which weights low frequencies (< 300 Hz) more strongly in the sound reduction factor, SF_6 no longer has any benefit over argon. This is the core of the statement that admixture of SF_6 to argon "leads to values that are independent of the type of gas fill" (Koch 1998).

A number of suppliers of soundproof glazing (e.g. Glas-Trösch) and of window systems (e.g. Unilux) no longer use SF_6 for the glazing - the former since 1992 and the latter since 1996.

²² The resulting possible 20-30% extra costs of the soundproof glazing only amount to a maximum of 5% extra cost for the whole window (including frame, fittings etc.), because the glazing only accounts for approx. 10 to 20% of overall window costs (Eschbacher 1999).



In Scenario II it is assumed that the use of SF_6 for soundproof windows is gradually phased out by 2005. Diagram 26 and Table 45 show the altered emissions.

Diagram 26: 1995-2010 SF₆ emissions from soundproof windows in Scenario II [t/a]. The main difference to Scenario I is the drop in overfilling (black area) to zero by the year 2005. This reduction potential (difference to Scenario I) in total emissions (uppermost white area) already amounts to 42 t in the year 2010.

- Over the 1995-2005 period, filling emissions (black area) drop to zero.
- From 2000 onwards, stock leakage losses drop slightly from 20 t to 17 t in 2010. In Scenario I they rise over the same period from 20 t to 23 t.
- Disposal losses develop in the same way as in Scenario I. They grow from 3 t in 2000 to 66 t in 2010, then forming the bulk of total emissions.
- Total emissions do not rise to 126 t until 2010 as in Scenario I, but primarily because of the absence of filling emissions only to 84 t.
- The reduction in total emissions compared to Scenario I (in Diagram 26 the uppermost white area) rises from 2000 to 2010 from 3 t to 42 t.

Table 45: 1995-2010 SF₀ emissions from soundproof windows						
in Scenario II [t SF ₆]						
1995 2000 2005 2010						
1. Filling losses	92	31	0	0		
2. Stock leakage losses	16	20	20	17		
3. Disposal losses	-	3	31	66		
Total emissions108545184						
Reduction potential	0	6	39	42		

5 Comparison of Global Warming Contributions of Scenarios I and II

The main difference between Scenario II and Scenario I is the gradual phase-out in the former of SF_6 filling by the year 2005. This measure immediately reduces filling losses, and also stock leakage losses over the medium term. However, as disposal losses continue to grow unchanged until 2020, the difference in terms of total emissions between the two scenarios is limited.

Table 46 and Diagram 27 show the global warming contributions of total emissions.

Table 46: 1995-2010 total SF₀ emissions from soundproof windows in Scenarios I and II [million t CO₂ equivalent]				
	1995	2000	2005	2010
Scenario I	2.57	1.43	2.14	3.02
Scenario II	2.57	1.28	1.21	2.01

In 2000, the first year of gradual phase-out of SF₆ filling, the emission reduction in Scenario II compared to Scenario I only figures 0.15 million t CO₂ equivalent. The reduction rises by 2010 to approx. 1 million t, at which time emissions from soundproof windows figure in Scenario I some 3 million and in Scenario II some 2 million t CO₂ equivalent.



Diagram 27: Global warming contribution of total SF_6 emissions from soundproof windows in Scenarios I and II. In 1999, the first year of SF_6 filling phase-out, the emission levels of the two scenarios separate. The global warming contribution in the year 2010 is more than 3.0 million t CO_2 equivalent in Scenario I, and only 2.0 million t in Scenario II.

3 Car tires

In 1995, car tires were the third largest SF_6 consumption sector in Germany after switchgear and soundproof windows. They were the largest emitter until 1998. From 1999 onwards, SF_6 emissions from this application drop substantially, both in the emissions forecast in Scenario I and in the reduction Scenario II. With a delay of 3 years, when the worn tires are decommissioned, the SF_6 escapes completely to the atmosphere. In this application, the utility is so out of proportion to the ecological damage that it causes that its continuation is extremely problematic.

1 Improved Pressure Stability through SF₆

Since 1984, car drivers in Germany can have their tires filled with SF_6 instead of air. The filling per tire is 250-350 g, i.e. for a set of 4 tires somewhat over 1 kg. The SF_6 molecules, being larger than those of air, diffuse much more slowly through the rubber. The inventor of SF_6 as tire filling gas (Continental AG) thus promised "stable tire pressure for 1 year and longer" (Continental, no year).

Constantly correct tire pressure contributes to improved driving safety, lower tire attrition and lower fuel consumption. According to Continental (ibid.), "more than half of all car drivers" are driving with pressure deviations from the design value. The system offered under the name of "Air Safe" can prevent upward and downward deviations from the correct filling pressure over almost the entire service life of a tire, as long as it is not made leaky by mechanical damage or defects in the wheel rim or valves.

This, however, is precisely a part of the problem. In practice, SF_6 filling can lead to neglect of regular pressure inspection (through placing the pressure gauge on the valve during air filling). In addition to safety concerns, there are also considerable ecological concerns. With a delay of three years after filling, when the worn tire is disposed of, the SF_6 banked in it is released completely to the atmosphere.

2 1995-1998 Consumption and Emissions in Germany

In 1995, the year of peak consumption, it was possible at more than 500 of the roughly 3500 retail outlets of the German tire trade to have tires filled with SF_6 gas. Tire traders offering SF_6 belonged to various retail chains or co-operatives supplied by the gas trade. SF_6 consumption for car tires in Germany rose from 1992 to 1995 from 110 to 125 t (Gas Trade Survey). 125 t SF_6 sufficed to fill more than 100,000 tire sets. This corresponds to 0.25% of the car fleet in Germany.

Emissions follow consumption with a delay of approx. 3 years. With an average car mileage of 15,000 km per annum and an average tire travel of 50,000 km, tire change is due every three years (Continental 1996a). When the tires are changed, the filling gas is released completely, be it SF₆ or air. The SF₆ consumption levels of the year 1992 are therefore the emissions of the year 1995, and in 1998 what was filled in 1995 is emitted etc. (cf. Diagram 28 and the data table therein).



Diagram 28: Car tires: 1995-1998 SF₆ consumption and emissions in t SF₆ and million t CO₂ equivalent. Annual SF₆ consumption (white columns) dropped over the 1995-1998 period from 125 to 30 t. However, due to the three-year delay, emissions rose over the same period (black columns). The emissions of 1998 were the consumed quantities of 1995. The global warming contribution (3rd line of the Table) of 1998 emissions figured 2.99 million t CO₂ equivalent.

Diagram 28 and the data table therein show a clear trend. Consumption (in the diagram the left, white columns) dropped over the 1995-1998 period from 125 to 30 t. However, due to the three-year delay, emissions still continued to rise over that period from 100 t to 125 t. (The 125 t emissions of the year 1998 are identical to the 125 t consumption of the year 1995.)

Emissions expressed in million t CO_2 equivalent rose over the 1995-1998 period from 2.63 to 2.99 million t (data table in Diagram 28).

The development for the three years after 1998 is predetermined: emissions drop to 30 t by 2001. This is the consumption of the year 1998.

3 Safety Concerns and Ecological Arguments against SF₆ in Tires

 SF_6 tire filling began to decline in 1996. In mid-1996, Continental ceased to directly supply the tire trade with SF_6 . However, Continental left it up to the tire traders to continue to satisfy customer demand for SF_6 , but no longer under the Conti Air Safe brand name (Glück 1999). The general gas trade supplied the gas. By 1998, further tire trade chains, including Stinnes Reifendienst (Petz 1999), had dispensed with SF_6 and offered nitrogen as an alternative. Nitrogen has been offered since 1996 instead of SF_6 by several other trader groups as a new tire filling gas for those car drivers who find air too common. In the meantime, some 1000 tire traders have nitrogen flasks for tire filling.

It is the behaviour of the customer group itself that gives rise to <u>safety concerns</u> over SF_6 gas as tire filling. These are not so-called sports drivers who devote particular attention to their vehicle. Most users of SF_6 value convenience and driving comfort, and have vehicles in the upper outfit and price categories (Continental 1996a). This purchaser group is further motivated by the claim that SF_6 in tires contributes to reduced driving noise and vibrations. Indeed, customers with SF_6 -filled tires regularly tell their dealers that they have a subjective feeling of improved driving comfort.

The official opinion delivered by Continental (see box) on this issue gives little credence to the objectivity of such statements (Continental 1996b). It may be that psychological factors play a role which accompany a commodity for which money has been spent (here 100 DM per set of four tires).

Consumption of and arguments for SF₆ in tires

Official opinion delivered by Continental AG on 21.10.1996

- 1. SF₆ in tires has a considerably lower diffusion rate than air and thus provides a longer constancy of air pressure over time, which permits optimum rolling resistance and minimized wear. However, this does not absolve from the need for regular air pressure monitoring, because when tires are damaged a gradual pressure loss can result which, in addition to safety aspects, also nullifies the above benefits.
- 2. The claim of customers that they experience improved comfort when using Air Safe could not be confirmed by test drivers on our testing site. However, we have not carried out any scientific study of this issue.

Considering the known impacts of SF_6 upon global warming as compared to the above benefits, Continental has discontinued its sales of SF_6 . However, we have left our trading companies free to continue to meet customer demand.

The <u>ecological</u> critique of SF₆ tire filling accepts that if inflation pressure is reduced by 0.3 bar compared to design pressure this leads to additional fuel consumption of 1.5%. Over an average lifetime of three years, this means about 60 litres more petrol. The combustion of this petrol generates about 140 kg CO₂. Avoiding this through correct tire pressure is certainly appropriate, but not if this is done by means of SF₆. If we compare the 140 kg CO₂ to the emission impact of 1 kg SF₆ after three years of tire service, namely 23,900 kg CO₂ equivalent, then it is all too evident that this is out of proportion. The global warming contribution of the SF₆ filling is almost 170 times larger than the reduction offered through less fuel consumption.

In view of this disproportion, SF_6 is not ecologically acceptable as a tire filling gas. Dispensing with it has no penalties in terms of practical usage, as SF_6 does not have the purpose of reducing the frequency of tire pressure measurements, but only the frequency of gas refilling. Who, however, has already placed the pressure gauge upon the opened valve to measure the pressure, will experience little inconvenience in spending a few extra seconds to press the knob to refill air.

4 2010 Business-as-Usual Emissions Forecast - Scenario I

Scenario I is not based on a renaissance of SF_6 in car tires, but on stabilization over the medium term of the consumption level reached in 1998. SF_6 is no longer recommended by tire manufacturers, nor marketed aggressively by retail chains. Nonetheless, the individual tire trader, whether member of a chain or independent, can continue to use SF_6 if the customers so desire. The customer pays for the SF_6 filling of a set of four tires approx. 100 DM, for nitrogen filling only 20 DM.

If the consumption level of 1998 remains constant until 2007, namely 30 t per annum, emissions drop by 2001 to this value, where they remain until 2010 (cf. Diagram 29).

5 2010 Relative Emission Reductions - Scenario II

Scenario II assumes that SF_6 is no longer used over the medium term as a tire gas. No statutory ban of this application is assumed, but an information policy pointing out the environmental damage caused by SF_6 tire filling. It is further expected of the tire manufacturers that they participate in awareness-raising efforts directed not only to car drivers but also to tire dealers. There is no advertising for SF_6 as tire gas, so that no new purchaser groups emerge.



Diagram 29: 1995-2010 emissions of SF_6 as tire filling gas in Scenarios I and II [t SF_6]. The drop in consumption since 1995 leads from 1999 onwards to declining emissions until 2001. From then on, emissions remain constant in Scenario I at 30 t per annum. In Scenario II (black area), emissions continue to drop, reaching zero in 2010.

Under these conditions, a further gradual but continuous drop in consumption and emissions is to be expected. It is assumed that consumption reaches zero in 2007, so that by 2010 at the latest there are no emissions any more.

6 Global Warming Contribution of SF₆ in Car Tires

Table 47: 1995-2010 SF ₆ emissions from car tires					
IN t SF	₆ and in mi		equivalent		
GWP: 23,900	1995	1998	2000	2005	2010
Scenario I emissions [t SF ₆]	110	125	50	30	30
Scenario II emissions [t SF ₆]				17	0
Million t CO ₂ equivalent	2.63	2.99	1.20	0.72	0.72
				0.40	0.00

The global warming contribution of SF₆ emissions from car tires figured approx. 2.63 million t CO₂ equivalent in 1995. In 1998, three years after the peak consumption year 1995, emissions reached the record level of 2.99 million t CO₂ equiv. (cf. Table 47). Emissions then drop, in Scenario I to 0.72 million t CO₂ equiv., in Scenario II to zero.

7 SF₆ Alternatives

Air is <u>the</u> alternative to SF_6 tire filling. However, several dealer groups advertise tire filling with nitrogen with the same argumentation as for SF_6 . Brochures promise "reduced tread wear and reduced pressure loss", because the gas "improves pressure stability", as it remains longer in the tire than "common air". This is the argumentation presented by "point S" and "Reifenring" and others in 1996.

It has not been possible to find measurement results giving an indication of the extent to which or indeed whether at all air, with its 20% nitrogen content, diffuses differently through rubber tires than 100% nitrogen. Pure nitrogen, as opposed to SF_6 , certainly makes no direct contribution to global warming. However, an indirect contribution does arise through the energy consumed for its production and transport, which is why the sometimes encountered designation "green" or "environmentally sound" tire gas is only appropriate in comparison to SF_6 , but not in comparison to air.

That pure nitrogen is used to fill aircraft tires is not indicative of its usefulness in cars. In aircraft, the tire filling must be, firstly, absolutely incombustible, and, secondly, absolutely dry in order to prevent corrosive condensation resulting from extreme temperature differences (Lufthansa 1996).

4 Magnesium Casting

In magnesium casting, SF_6 is used as a protective cover gas over the molten metal in order to prevent oxidation and burning. It is distributed at concentrations of 1 vol.-% in blends with carrier gases such as dry air or CO_2 over the surface of the more than 650°C hot molten metal, which would otherwise ignite at temperatures over 500°C (Solvay 1992, 16/17). The SF_6 supplied²³ only undergoes minimum chemical conversion, so that consumption and emissions can be equated.²⁴

 SF_6 has been used in this application since the mid 1970s, then competing with SO_2 , which in concentrations of 0.5-1.5% over the melt forms a protective layer of MgSO₄ (Gjestland/Westengen 1996, 7). As SF_6 is simpler to handle than the highly toxic SO_2 , it became the cover gas of choice in many new casting companies.

Cover gas is used in Germany only to process magnesium which is imported in ingots. When casting the freshly produced metal in these ingots, cover gas is also used in the primary producer countries, and in larger amounts than in German casting companies. In the following, the SF₆ used abroad in primary production is not considered, but only SF₆ usage within Germany.

1 1995-1997 Consumption and Emissions in Germany

Over the 1995-1997 period, the use in Germany of SF_6 as cover gas dropped from 6.1 to 3.0 t (cf. data table of Diagram 30), although magnesium casting production rose over the same period from 5000 to 12,000 t.

In 1995, the ca. 20 German magnesium casting companies produced about 5000 t cast magnesium parts. SF_6 was only used as cover gas for 2000 t Mg (40%), SF_6 consumption totalling 6.1 tonnes. The bulk of cast Mg products was produced with SO_2 . In the following three years, 1996, 1997 and 1998 (preliminary figures), magnesium product production leapt to three times the previous figures, namely 14,600 t in 1998. Over the same period, absolute consumption of SF_6 fell by half to 3.1 t (Magnesium Casting Survey 1999).

That SF₆ consumption has dropped while magnesium output has grown has two causes:

<u>Firstly</u>, casting companies which in 1995 still cast 1000 t magnesium using 3.4 t SF_6 switched to SO_2 . This is why the absolute consumption of SF_6 dropped by half.

<u>Secondly</u>, four new foundries were established which, while still using SF₆ and already producing in 1997 5500 t (1998: 7350 t) magnesium, were able to cut specific gas consumption per tonne magnesium produced from 2-3 kg by a factor of 100 to 25-30 g. This was made possible by new, SF₆-saving furnace technology (Magnesium Casting Survey). These four modern foundries, which belong to the car manufacturing industry and its component suppliers, consumed only 150 kg SF₆ for the 5500 t Mg in 1997.

 $^{^{23}}$ "SF₆ reacts with the surface of an Mg melt, forming a protective film which prevents further Mg reactions with atmospheric oxygen" (Gmelin 1978, 184).

 $^{^{24}}$ "In the magnesium industry all SF₆ used in production is released to the atmosphere." (Gjestland/Westengen/Magers 1996, 2).



Diagram 30: 1995-1998 SF₆ consumption for magnesium casting in t/a and kg per tonne Mg. Despite production trebling over this period, absolute SF₆ consumption (columns, left scale) dropped from 6.13 t (1995) to 3.13 t (1998). Thanks to the use of modern technology, average specific SF₆ consumption per tonne magnesium produced (line, right scale) dropped from 3.09 to 0.34 kg.

Owing to the annually growing importance of new as compared to old SF_6 furnace technology, over the 1995-1998 period the average emission coefficient over all foundries using SF_6 dropped from 3.09 to 0.34 kg SF_6 per tonne magnesium produced (cf. Diagram 30, data table, line 2).

In the year 1997, of a total of 12,000 t magnesium casting produced, 41% used SO₂ as cover gas, 13% used SF₆ with old furnace technology (2 kg/t Mg) and 46% used SF₆ with new furnace technology (27 g/t Mg). It was thus possible to increase several times over the absolute production of magnesium casting while at the same time emitting less SF₆ than in the year 1995.

SF₆ Savings Through New Furnace Technology

Specific SF₆ emissions are particularly high in small-scale discontinuous production, because warm-up and cooling phases extend the production time and the process of charging the crucible usually involves opening its hatch. Moreover, in order to remove iron fluoride scaling from the crucible walls, the lid needs to be opened manually. Opening the lid during operation is a major source of emissions, as the atmosphere over the hot melt surface has high buoyancy. In many cases the cover gas is supplied by manual control and by the eye, meaning when in doubt "preferably a bit more" in order to be "on the safe side" (Frech 1999).

Once the high global warming potential of SF_6 became known, magnesium suppliers warned their customers against overdosing (Hydro Magnesium 1996). But it is only since casting plant manufacturers have made SF_6 reduction their particular concern that specific SF_6 emissions have dropped significantly.

Emission abatement is based inter alia on the following technical measures:

- 1. The tightness of the ovens is improved. The refractory seal between the crucible and the lid prevents gas from escaping, and material charging is fully automatic and continuous, and passes through a lock system.
- 2. Cover gas dosage is fully automatic by electronic control of both gas mix and flow rate, and of supply pressure, in accordance with the needs indicated by the state of the melt (Rauch 1999; Westofen 1999).
- 3. Large foundries have the further benefit of three-shift operation, which minimizes warm-up and cooling losses (Scherelis 1996).

2 2010 Business-as-Usual Emissions Forecast - Scenario I

It is assumed in both emissions scenarios that production capacities grow from 1998 to 2010 by 20% annually through new plants (Hartmann 1999). Annual output rises to 73,000 t by 2010. New, modern, large-scale plants multiply their production from 7350 to 65,500 t over the 1998-2010 period.

It is mainly because of increased use in car manufacturing that such a strong growth in demand is expected for magnesium casting over the next years in Germany. This phase of capacity expansion already commenced in 1996 with the establishment of four new foundries. The light metal, with a density of 1.8, is used to substitute steel (density approx. 7.8) in order to reduce vehicle weight - initially in steering wheels, gearbox cases etc.

Scenario I makes the two following assumptions concerning cover gas:

- 1. All <u>new</u> plants use SF₆ with modern furnace technology. Specific gas consumption per t magnesium casting figures 30 g.
- 2. In the <u>old</u> plants, the already planned conversion of 1600 t capacity to SO₂ is implemented by 2000. The annual production of 7000 t (SO₂ technology) reached then is not further increased. From 2000 onwards, the old plants produce 7000 t magnesium casting with SO₂ and 600 t with SF₆. The 600 t are produced in seven small plants with discontinuous processes, in which specific SF₆ consumption per t magnesium casting is constant at 4.5 kg.

Under these conditions, the following SF₆ emissions arise until 2010:

From the initial empirical value of 3.12 t in the year 1998, emissions rise slowly until 2010 to a total of 4.67 t. Of this, the greater part, namely 2.7 t (58%), continues to be emitted by the small old plants with an annual output of 600 t magnesium product. The new large-scale plants produce in 2010, with 65,500 t, a good hundred times more than the old small plants, but only use a little less than 2 t SF₆ for this. In the business-as-usual Scenario, total SF₆ emissions in 2010 are still 24% below the value of 1995.

3 2010 Emission Reductions - Scenario II

The largest European magnesium producer, Norsk Hydro, fears for its product that "the use of SF₆ greatly reduces its attractivity" because the light metal, which, owing to fuel saved in car transport, permits reduced CO_2 emissions, loses its favourable global warming profile in an environmental life-cycle assessment due to too much SF₆ consumption in its production (Gjestland/Westengen 1996, 6). The authors also refer to the SF₆ used in primary casting in the producer country, citing current emissions of "0.5 kg per tonne magnesium casting". They support in principle conversion to SO₂: "The use of SF₆ in the magnesium industry should be reduced strictly or even eliminated" (ibid.). Norsk Hydro, specifically its Hydro Magnesium branch, recommends in principle that its clients use SO₂ as cover gas (Hartmann 1999). This has been successful among specialized light metal casting companies.

1995-2010 SF₆ emissions from magnesium casting in tonnes per annum - Scenarios I and II 7.0 6.0 5.0 4.0 Tonnes 3.0 Reduction potential 2.0 Scenario II 1.0 0.0 1995 2000 2005 2010

Reduction Scenario II assumes the same magnesium casting production until 2010 as Scenario I. However, the assumptions concerning the cover gas differ:

Diagram 31: 1995-2010 SF₆ emissions from magnesium casting in tonnes per annum - Scenarios I and II. Through using SO₂ instead of SF₆ as cover gas, SF₆ emissions drop by half over the 1995-2000 period. In Scenario I, despite magnesium casting output growing ten-fold, emissions rise only slightly until 2010 owing to SF₆-saving furnace technology. In Scenario II, they even continue to drop after 2000, because SF₆ is replaced throughout as cover gas, except for minor uses.

- 1. The small companies, with an annual output together of 600 t, also largely retrofit their processes from SF_6 to SO_2 from 2000 to 2010, leaving only 10% of their production using SF_6 .
- 2. From 2002 onwards, new large-scale foundries belonging to the car manufacturing or car component supplier industry also no longer protect the magnesium melt with SF_6 , but with SO_2 or another SF_6 substitute. From 2005 onwards, SF_6 is also substituted in the foundries that were newly established since 1996.

Under these conditions, SF_6 emissions are reduced compared to Scenario I from 2000 onwards. In the year 2010, they come exclusively from small companies in which 10% of production capacity has not been converted to a different cover gas. In Scenario II, total emissions in the year 2010 only figure 0.27 t (270 kg).

4 Comparison of the Emissions of the two Scenarios

Diagram 31 and Table 48 show the SF₆ emissions of the two scenarios. In the year 2010, the savings in Scenario II compared to Scenario I figure 4.4 t SF₆ or 0.105 million t CO_2 equivalent.

Table 48: 1995-2010 SF ₆ emissions from magnesium casting						
Scenarios I and II						
1995 2000 2005 2010						
Scenario I emissions [t SF ₆]	6.13	3.02	3.49	4.67		
Scenario II emissions [t SF ₆]			1.73	0.27		
Reduction potential [t SF ₆]	-	-	1.76	4.40		
Reduction in mill. t CO ₂ equiv.	-	-	0.042	0.105		

Diagram 32 documents graphically the comparative global warming contribution of the two scenarios. In Scenario II, emissions in 2010 are far below the initial value of 1995, but are not reached again in Scenario I, either.



Diagram 32: Global warming contribution of SF_6 emissions from magnesium casting -Scenarios I and II. Even in the business-as-usual Scenario I, the initial emissions of 1995 (0.146 million t CO₂ equivalent) are not reached again by 2010. In Scenario II (maximum SF_6 substitution) the global warming contribution only figures 0.006 million t CO₂ equivalent.

5 Semiconductor Manufacturing

In the semiconductor industry, SF_6 is an important gas for wafer etching in low-pressure plasma. This application of SF_6 is treated in Part IV of the present report together with other fluorinated gases used in semiconductor manufacturing. The following consumption and emission data are taken from that Part.

1 1995-1997 Consumption and Emissions in Germany

Table 49: 1995-1997 SF₀ consumption and emissions in the semiconductor industry					
1995 1996 1997					
Consumption [t SF ₆]	7	9	10		
Emissions [t SF ₆]	4	5	5		
Emissions in million t CO_2 equiv. 0.097 0.114 0.126					

The difference between consumption and emissions results firstly from the only partial chemical transformation in the plasma reactor, and secondly from the effect of downstream exhaust gas purification.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

The assumptions for Scenarios I and II are presented in Part IV. Here only the resultant emissions are presented.

Table 50: 1995-2010 SF₀ emissions from semiconductor manufacturing in business-as-usual Scenario I				
	1995	2000	2005	2010
Emissions [t SF ₆]	4	6	8	11
Million t CO ₂ equivalent	0.097	0.150	0.200	0.268

In business-as-usual Scenario I, the emissions shown in Table 50 result until 2010. SF_6 emissions rise over the 1995-2010 period from 4 t to 11 t.

3 2010 Emission Reductions - Scenario II

In Scenario II, lower emissions occur from 2000 onwards (Table 51). By the year 2010, they are 2 t below the initial value of 1995 (4 t).

Table 51: 1995-2010 SF ₆ emissions from semiconductor manufacturing in reduction Scenario II				
	1995	2000	2005	2010
Emissions [t SF ₆]	4	6	6	2
Million t CO2 equivalent	0.097	0.137	0.138	0.045

Scenario II provides an emissions reduction of 9 t in the year 2010. Instead of 0.268 million t CO_2 equivalent in Scenario I, the global warming contribution only figures 0.045 million t, being 17% of the former figure.

6 Other SF₆ Applications

In addition to the five main applications of SF_6 , there are a number of further individual applications which emit small quantities individually, but add up to emissions that are by no means negligible. Eight different SF_6 emission sectors have been identified, ranging between 70 kg/a and 3100 kg/a. Together, these already emitted 7.3 t in 1995 (cf. Table 52).

Table 52: 1995-1997 SF ₆ emissions in Germany from 'other applications' in t/a						
	1995	1996	1997			
Particle accelerators	2.9	3.1	3.1			
Electron microscopes	0.2	0.2	0.27			
Radio materiology	0.23	0.26	0.3			
Radiotherapy	0.07	0.08	0.09			
Tracer gas	0.5	0.5	0.5			
Military aircraft radar	1.0	1.0	1.0			
Aluminium cleaning	0.5	0.5	0.5			
Miscellaneous	2.0	2.0	2.0			
Total	20510	2049	7.76			

1 Tandem Particle Accelerator with Van-de-Graaff Generators

Nuclear research uses, among other types of equipment, electrostatic particle accelerators (cf. NuPECC 1995). In these tandem accelerators with Van-de-Graaff generators, the high voltage required for ion acceleration in the vacuum is applied externally. The equipment is placed together with the high-voltage generator in a tank filled with insulating gas. SF_6 is not always the gas used for insulation; where it is used, then it is either alone or in blends with other gases in varying concentrations. In order to be able to manipulate the vacuum tube for research, the tank needs to be emptied. The insulating gas is then pumped into a storage tank, cleaned or dried and subsequently pumped back again. Gas losses occur in pumping, and also when pressure relief valves are actuated etc., as the equipment is generally operated under relatively high pressure (in the cases examined between 5 and 25 bar).

Equipment sizes differ; in the examined cases, the SF_6 filling ranged from 10 kg to almost 16 tonnes. Annual emissions differ greatly, depending upon the size and operating conditions of the overall facility. 18 facilities at 14 sites (universities and public research institutions) were surveyed. The aggregation of the SF_6 stock and emissions shown in Table 53 follows the information provided by the facility operators. These determined their operating emissions from their annual refills. Annual emissions amount to about 7.5% of the stock.

Consumption for new facilities (not consumption for loss replacement) is discontinuous and is not taken into consideration here. Since 1965, one new facility has been installed every two years on average. Between 1990 and 1995, two facilities were commissioned, one in 1992 with 360 kg SF₆ and the other in 1993 with 900 kg SF₆. In 1996 two further facilities were installed, the two amounting to 1750 kg SF₆, so that the total SF₆ stock rose since 1995 by about 2 t, from 39 to 41 t.
Table 53: 1995-1997 SF₀ emissions from particle accelerators in t/a								
Year	ear 1995 1996 1997							
Emissions [t/a]	2.9	3.1	3.1					
Stock [t] 39 41 41								
Osumes Deutiste Asselsu	- (

Source: Particle Accelerator Survey; stock emission rate: 7.5%.

2010 Emissions forecast

It is to be expected that the stock does not grow, but remains roughly constant. <u>Stock</u> <u>emissions</u> therefore remain at a level of 3.1 t/a over the long term (until 2010). It is assumed that when equipment is decommissioned in the future, the SF₆ is recovered and reprocessed and that there are therefore no scrapping emissions (Solvay 1996).

2 High-Voltage Electron Microscopes

In a small percentage (< 10%) of the transmission-type electron microscopes installed in Germany, SF₆ is used to insulate the high-voltage (HV) generator and emission chamber. Such insulation is not necessary for units in the voltage range below 200 kV. Since they were first introduced in the mid-1980s, the stock of SF₆-insulated units in the 200-400 kV voltage range has grown annually by some 6 units. The leading supplier in Germany estimates the stock at 60 in 1995 and more than 70 units in 1997 (Fei 1999). The quantity of SF₆ contained in these electron microscopes ranges from 8 kg in 200 kV units to 40 kg in 400 kV units, on average about 18 kg. The total SF₆ stock in units installed in Germany amounted to 1080 kg in 1995 and 1300 kg in 1997.

The units have a lifetime of 15 years, and annual leakage rates of about 0.5 kg, regardless of the type of unit (Fei 1999). Repairs are usually carried out at the manufacturer. However, according to statements of users, on-site emissions in the event of accident and in the course of repairs must also be expected, which are integrated in the figures given in Table 54 at a level of a further 0.5 kg per unit. Annual emissions (refills) thus figure 1 kg per unit, or a total of 60 kg in 1995 and 70 kg in 1997. These figures do not include emissions arising when old units are decommissioned, as their return to the manufacturer and recovery of the SF₆ is assumed (Fei 1999).

In addition to electron microscopes in the kilovolt range, there are also SF_6 -insulated large units in the megavolt range (> 1000 kV), which account for some two thirds of the overall SF_6 stock. There are presently three of these in Germany, together totalling some 2000 kg SF_6 . Over the 1995-1997 period, one such unit with a filling of approx. 500 kg was newly installed. Annual SF_6 consumption for emissions compensation is calculated by users at approx. 10% (Max-Planck-Institut 1996), this meaning 200 kg/a.

Table 54: 1995-1997 SF₀ emissions from HV electron microscopes in kg/a							
	1995 1996 1997						
Kilovolt units	60	65	70				
Megavolt units	140	140	200				
Total	200	205	270				

Table 54 compiles the continuous emissions from the stock of kilovolt and megavolt units. These rose in total over the 1995-1997 period from 200 to 270 kg.

2010 Emissions forecast

Annual emissions (refills) for kilovolt units will rise to 100 kg by the year 2000. Because in that year saturation of the stock (100 units) is expected, emissions will presumably not continue to rise. Forecasts are difficult for megavolt units, because only three of these were installed over the past 25 years. No growth in emissions is expected here. If the SF₆ recovery concept for electrical equipment (discussed in Section 1 in this Part of the present study) is implemented and recovered gas from old equipment is available for refilling, no substantial growth of either emissions or new consumption is to be expected.

It is estimated that in 2010 annual emissions will total 300 kg.

3 Radio Materiology

In portable radio materiology units (for weld seam testing etc.), SF_6 is preferred instead of the otherwise usual oil insulation for weight reasons. Since the late 1980s, the stock of these units has grown continuously. It is estimated by manufacturers (Philips Industrieröntgen 1996; Yxlon 1999) as follows: 1995 ca. 500, 1997 ca. 700 units. Over the 1995-1997 period, annual sales in Germany are estimated at about 100 units. With an average filling of 1.5 kg SF₆, this results in an annual growth of the SF₆ stock of 150 kg. In 1995, the stock amounted to about 750 kg, in 1997 to about 1050 kg.

Gas losses arise when new units are filled and after repairs (15% of refill) and maintenance (20% of the stock annually).

About 250 new units are produced annually, most of which are exported. When new units are filled, filling losses of ca. 15% occur, amounting to about 55 kg annually.

About 20% of the stock in Germany is repaired each year. This involves the release of the SF₆. The refills for these gas losses amounted to 150 kg in 1995 and 210 kg in 1997. In addition to these repair refills, there were 15% filling losses: ca. 20 kg in 1995, ca. 30 kg in 1997.

Table 55 shows the total emissions of 1995-1997.

Table 55: 1995-1997 emissions from radio materiology units in kg/a:						
Year	1995	1996	1997			
Emissions [kg/a]	230	260	300			

2010 Emissions forecast

In Germany, saturation of the radio materiology unit market is assumed for the year 2000, with a stock of 1000 units (1500 kg SF_6 filling). The lifetime is reported to be at least 10 years. SF_6 -insulated units have been on the market since the late 1980s.

Disposal emissions therefore also play a role from 2000 onwards, in addition to filling and maintenance losses. A disposal rate of 100 units per year is assumed. The 150 kg released in disposal are not recovered. In the state of saturation after 2000, filling losses for new units amount to 55 kg annually, filling after repairs to 300 kg (=20% of the stock), and the filling losses associated with repair fillings 45 kg. To this is added 150 kg emissions in disposal.

These total emissions of 550 kg per annum are expected to remain stable until 2010.

4 Radiotherapy (Linear Accelerators)

In these devices used for medical purposes, the SF_6 filling per unit is small - slightly more than 0.1 kg. It is generally exchanged by the user four times a year, which corresponds to just under 0.5 kg SF_6 annual consumption - and annual emission - per unit. The gas is not at present captured. Recovery is not planned.

As SF₆ is only in use as an insulating gas since 1992 (Elekta 1999), the equipment stock is at present still small. 140 SF₆-insulated units can be assumed for 1995, with annual growth of 15 units. Refills for emissions compensation increased over the 1995-1997 period from 70 to 85 kg, and are expected to figure 110 kg in the year 2000.

Table 56: 1995-1997 SF₀ emissions from radiotherapy units in kg/a					
Year	1995	1996	1997		
Emissions [kg/a] 70 77 85					

2010 Emissions forecast

Equipment saturation is expected from about 2005 onwards. The stock will then figure about 300 units, requiring 150 kg annually for emissions compensation. Unit lifetime is estimated at 12 to 15 years (Elekta 1999).

Disposal of old equipment, which will set in from 2000 onwards, is expected to involve low emissions of less than 0.1 kg per unit, which are not taken into consideration here.

5 Tracer Gas for the Study of Air Flows

As SF₆ is a trace gas that is both stable and readily detectable even in extremely low concentrations, it is used to study ground-level and atmospheric air flows and gas dispersal (Zenger et al. 1994). Experts estimate that max. 100 kg/year are used for urban climate and siting studies (cold air dispersal, fresh air passages; landfill and sewage plant design etc.).

The research sector (large-scale research centres) consumes 250-400 kg annually. Here consumption and emissions are identical. Total emissions can be estimated at 400-500 kg SF_6 annually (Tracer Gas Survey).

Table 57: 1995-1997 emissions of SF ₆ as tracer gas in kg/a						
Year	1995	1996	1997			
Emissions [kg/a]	500	500	500			

2010 Emissions forecast

There are no indications at present that past usage of 500 kg per annum will change in the future.

6 Military Aircraft Radar

 SF_6 is used as an insulating medium in the radar systems of the large AWACS military reconnaissance planes. Its purpose is to prevent electric flashovers in the hollow conductors to the antenna, in which high voltages of more than 135 kV prevail (Dornier 1996). All other radar systems for aircraft in Germany, be it ground or aircraft radar, primary or passive, are operated at lower voltages (up to 30 kV), so that no SF_6 is necessary, oil (silicone oil) sufficing (Dasa 1996). AWACS systems are, strictly speaking, not aircraft radar systems, but rather ground radar stations built into aircraft, which must operate at high power while needing little space.

In the maintenance of the AWACS fleet of NATO, about 1000 kg SF_6 are consumed annually as emissions replacement (Dasa 1999).

Table 58: 1995-1997 SF₀ emissions from AWACS radar in kg/a						
Year 1995 1996 1997						
Emissions [kg/a] 1000 1000 1000						

2010 Emissions forecast

As AWACS aircraft are to be retained over the next twenty years, no change in SF_6 emissions is to be expected.

7 Cleaning Gas for Secondary Aluminium Casting

In aluminium smelters, the inert gases nitrogen and/or argon are supplied prior to casting in order to remove (degas) hydrogen, alkali metals and alkaline earth metals and solids. This prevents porosity occurring in the castings. While elementary chlorine is usually added to the inert gases in primary melt cleaning because purity requirements upon primary aluminium are very high, in secondary aluminium smelting inert gases without additives generally suffice (Steinhäuser 1996).

Table 59: 1995-1997 SF₀ cleaning gas in aluminium melting in kg/a							
Year	1995 1996 1997						
Emissions [kg/a] 500 500 500							

In a small number of - usually smaller - secondary aluminium smelters and in laboratories, an inert gas cleaning system is used to which SF_6 is added in concentrations of 1 or 2.5% (AGA-Gas 1999; Westfalen AG 1999).

The gas trade estimates that the SF_6 used in Germany for this purpose amounts to 500 kg/a - with a constant, by no means rising market development since 1990.

2010 Emissions forecast

There is at present no alternative to this gas mixture, so that the same SF_6 quantity is to be expected for 2005 and 2010.

8 Miscellaneous Applications, Including Filling Gas in Sports Shoes

The research conducted in connection with the present study revealed a number of very small SF₆ consumers, such as mass separators, Czerenkov detectors etc., which were not surveyed precisely because the quantities used are marginal in each application and only noteworthy when taken together. A larger individual usage of SF₆ was notified by an American sport attributes company, with the request to maintain confidentiality. This is SF₆ in the soles of sports shoes ("Air System") imported to Germany (Nike 1996).

If we add up the unidentified, and consequently only estimated, SF_6 quantities with the SF_6 quantities subject to discretion, then total emissions of approx. 2 to 2.2 t SF_6 annually are a realistic quantity for the 1995-1997 period. Concerning shoe sole filling, it needs to be noted that the SF_6 is emitted in Germany, but the shoes are manufactured abroad.

Table 60: 1995-1997 SF₀ emissions from miscellaneous areas, including sports shoes, in t/a						
Year	ear 1995 1996 1997					
Emissions [t/a] 2.0 2.1 2.2						

Sources: own estimate; Nike 1996; sports shoe filling is not listed separately.

2010 Emissions forecast

It can be expected that the total emissions of all miscellaneous areas incl. sports shoes will not increase after 2000, but will stabilize at 2 t. This is because Nike has decided to phase out the use of SF_6 in shoe soles, in favour of nitrogen. SF_6 phase-out in new filling shall be concluded by 2001 (Nike 1999). From 2005 onwards, no SF_6 emissions shall occur in Germany any more from sports shoes, whose lifetime is estimated at 3 to 4 years.

9 Presumed but not Verified SF₆ Applications

A difference of approx. 8 t per year was found between the total of the annual sales figures reported by SF_6 producers and gas traders on the one hand and the consumption figures reported by users in Germany on the other. This gave reason to search for further SF_6 applications. This research did not find any further noteworthy

areas of application. In Germany, SF_6 is not used in laser equipment (Poprawe 1996), not in tennis balls (Dunlopillo 1996) and evidently also not as leak detection gas.

10 1995-1997 Global Warming Contribution

Multiplication of the total annual emissions from the eight 'other' application sub-sectors by the GWP of SF_6 leads to a global warming contribution of 0.177 to 0.185 million t CO_2 equivalent over the 1995-1997 period.

Table 61: SF ₆ emissions from 'other' applications in t SF ₆ and in million t CO₂ equivalent							
GWP: 23900 1995 1996 1997							
Emissions [t SF ₆]	7.4	7.64	7.76				
Mill. t CO2 equivalent	Mill. t CO_2 equivalent0.1770.1830.185						

11 2010 Total Emissions Forecast for 'Other' Applications

In the above text of Section 6, emissions forecasts until 2010 were given for each of the eight sub-sectors in addition to the emissions estimates for the 1995-1997 period. These are listed and totalled in Table 62.

The totalled emissions forecast extrapolates present usage trends. It can be qualified as a business-as-usual forecast. However, it is already based on the assumption that the concept for reusing spent SF_6 gas from electrical equipment (cf. Section 1) is implemented in particle accelerators and in radio materiology. It thus makes little sense to elaborate a second scenario forecasting substantial emissions reductions, as there is presently no basis to assume that these will take place.

Table 62: 1995-2010 SF ₆ emissions in Germany from 'other' applications								
in t/a SF ₆ and million t CO ₂ equivalent								
	1995	2000	2005	2010				
Particle accelerators	2.9	3.1	3.1	3.1				
Electron microscopes	0.2	0.3	0.3	0.3				
Radio materiology	0.23	0.55	0.55	0.55				
Radiotherapy	0.07	0.11	0.15	0.15				
Tracer gas	0.5	0.5	0.5	0.5				
Military aircraft radar	1.0	1.0	1.0	1.0				
Aluminium cleaning	0.5	0.5	0.5	0.5				
Miscellaneous	2.0	2.0	2.0	2.0				
Total	7.40	8.06	8.10	8.10				
Ems. in mill. t CO2 equiv.	0.177	0.193	0.194	0.194				

In total, SF_6 emissions from 'other' applications are anticipated to rise over the 1995-2010 period from 7.40 t to 8.10 t.

The global warming contribution thus rises over that period from 0.177 to 0.194 million t CO_2 equivalent, or by 10%.

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III. Perfluorocarbons (PFCs)

Overview

 CF_4 and C_2F_6 are the trace gases in the group of perfluorocarbons (PFCs) which have the greatest quantitative global warming impacts. Natural sources have been identified for CF_4 in the atmosphere in addition to anthropogenic sources (Harnisch/Eisenhauer 1998). No indications have yet been found of a natural source of C_2F_6 (Harnisch 1996). The growth in global atmospheric CF_4 concentrations beyond the natural background concentration that has been observed since the mid-1930s correlates with the worldwide growth in primary aluminium production (ibid.), this being viewed as the main source of anthropogenic emissions of CF_4 and of C_2F_6 .²⁵ In Germany, too, aluminium production is the largest emitter of CF_4 . However, since 1996 the semiconductor industry is the largest emitter of C_2F_6 (and the second largest emitter of CF_4). C_3F_8 is the only longer-chain PFC with a noteworthy contribution to emissions in Germany.

In the following, two scenarios are elaborated for each application sector for the PFC emissions from the territory of Germany until the year 2010.

Scenario I is a business-as-usual scenario and extrapolates present trends largely unaltered. Scenario II is a reduction scenario in which existing technological potentials to reduce emissions or to substitute PFC usage are exploited in each sector.

In Aggregate Scenario I - the total of the four sectoral business-as-usual scenarios -PFC emissions rise over the 1995-2010 period from 258 t to 340 t. In Aggregate Scenario II, they drop to 167 t. In the year 2010, emissions in Scenario II are only about half those of Scenario I. The global warming contribution in 2010 figures some 2.52 million t CO_2 equivalent in Scenario I and 1.15 million t in Scenario II. This means a reduction in Scenario II as compared to Scenario I by 1.37 million t CO_2 equivalent, or by 54%.

1 2010 Business-as-Usual Emissions Forecast - Scenario I

Business-as-usual Scenario I is based upon the following sectoral assumptions for the 1995-2010 period.

- In <u>aluminium electrolysis</u> the production capacity of smelters in Germany remains constant at the level of 1998. The technology breakdown of total capacity in 1998 between electrolysis cells with alumina feeding from the side (SWPB) and centre point feeding (PFPB) also remains as in 1998. Both feeding technologies become 1% more efficient each year.
- In the semiconductor industry 95% of chamber cleaning is conducted using PFCs (C₂F₆, CF₄ and C₃F₈) in the first ten years after 2000, as in the 1990s. Emissions grow in step with the anticipated above-average consumption growth of 11% per year for chamber cleaning, saturation with exhaust gas systems for PFC destruction remaining equal.

 $^{^{25}}$ C₃F₈ emissions from aluminium smelters are lower by an order of magnitude, figuring 0.3% of CF₄ emissions (Harnisch 1996, 58). The present study does not consider C₃F₈ from aluminium smelters.

- 3. Annual consumption (4 t) and emissions (3.4 t) of CF₄ in <u>printed circuit board</u> manufacture remain constant.
- 4. <u>Refrigerant</u> blends containing PFCs (C_3F_8 , C_2F_6) are only used to substitute CFCs, but not HCFCs in old equipment. Their new consumption terminates in the year 2000, their emissions end in the year 2005.

Under these assumptions, the PFC emissions shown in Table 63 result for all sectors together over the 1995-2010 period. The total rises from 258 t to 340 t (last line), this rise being almost completely attributable to the steep rise in emissions from semiconductor manufacturing. In that industry, CF_4 emissions rise over the 1995-2010 period by 65 t (line 2) and C_2F_6 emissions by 90 t (line 6). PFC emissions from aluminium smelters drop over the same period by 67 t CF_4 and 7 t C_2F_6 (lines 1 and 5).

Table 63: 1995-2010 PFC emissions by source							
in b	ousiness-	as-usual So	cenario I [t/	aj			
Source	PFC	1995	2000	2005	2010		
1. Aluminium smelters	CF_4	209	157	149	142		
2. Semiconductor man.	CF_4	12	31	50	77		
3. Printed board man.	CF_4	3.4	3.4	3.4	3.4		
Sub-total	CF₄	2223	2195	2211	2236		
5. Aluminium smelters	C_2F_6	21	16	15	14		
6. Semiconductor man.	C_2F_6	11	36	62	101		
Sub-total	C_2F_6	4478	4442	4499	4587		
7. Semiconductor man.	C_3F_8	0	1	2	3		
8. Refrigerant blends	C₃F ₈	2	15	0.03	0		
Sub-total	C₃F ₈	2	16	2	3		
Total of all PFCs	Total of all PFCs 258 259 281 340						

In comparison to CF_4 and C_2F_6 emissions from semiconductor manufacturing and aluminium electrolysis, the other PFC emissions are of subordinate importance. CF_4 emissions from printed circuit board manufacture (Table 63, line 3) remain constant (as assumed) at the low level of 3.4 t. C_3F_8 emissions from refrigeration (line 8) drop to zero by 2010. C_3F_8 emissions from semiconductor manufacturing (line 7) grow in relative terms, but are low in absolute terms.

Table 64: 1995-2010 PFC emissions by source in business-as-usual Scenario I in million t CO₂ equivalent						
Source 1995 2000 2005 2010						
1. Aluminium smelters	1.551	1.165	1.108	1.054		
2. Semiconductor man.	0.174	0.542	0.904	1.445		
3. Printed board man.	0.022	0.022	0.022	0.022		
4. Refrigerant blends 0.015 0.103 0 0						
Total	3.757	3.832	4.039	4.531		

Table 64 lists the global warming contributions of sectoral PFC emissions. Total emissions in the year 2010 amount to 2.52 million t CO₂ equivalent. 57% of this is attributable to semiconductor manufacturing and 42% to aluminium smelters.

2 2010 Emissions Reduction - Scenario II

The reduction scenario (Scenario II) has the following assumptions for 1995-2010:

- In <u>aluminium electrolysis</u> the production capacity of smelters in Germany and the annual 1% efficiency increase in both alumina feeding technologies remain the same as in Scenario I. The difference lies solely in the assumption that the remaining cells with side feeding (SWPB) are retrofitted to centre point feeding (PFPB), so that by 2010 all of aluminium production in Germany uses the most modern and least PFC-generating technology.
- 2. In the <u>semiconductor manufacturing</u> industry, a revolution in chamber cleaning technology sets in from the year 2000 onwards. In newly installed facilities, PFC is no longer used, but only the fluorinated, but carbon-free gas NF₃. As 98-99% of the NF₃ is chemically decomposed before entering the process and does not recombine, not only are there no longer any emissions of PFCs (CF₄, C₂F₆, C₃F₈) but there are also only slight NF₃ emissions in their place.
- 3. In the <u>printed circuit board manufacture</u> and <u>refrigeration</u> sectors, no emissionabating assumptions are made. Here PFC emissions are the same in both aggregate scenarios.

Table 65: 1995-2010 PFC emissions by source					
in reduction Scenario II [t/a]					
Source	PFC	1995	2000	2005	2010
1. Aluminium smelters	CF ₄	209	157	131	124
2. Semiconductor man.	CF_4	12	27	27	11
3. Printed board man.	CF ₄	3.4	3.4	3.4	3.4
Sub-total	CF₄	224	188	161	139
5. Aluminium smelters	C_2F_6	21	16	13	12
6. Semiconductor man.	C_2F_6	11	31	28	11
Sub-total	C_2F_6	2479	47	41	23
7. Semiconductor man.	$C_{3}F_{8}$	0	1	2	3
8. Refrigerant blends	C₃F ₈	2	15	0.03	0
Sub-total	C ₃ F ₈	2	16	2	3
Total of all PFCs		258	250	204	165

Under these conditions, the lower PFC emissions shown in Table 65 result for all sectors together over the 1995-2010 period.

Total PFC emissions drop from 1995 to 2010 from 258 t to 165 t. Compared to the 1995 baseline, this is a reduction by 93 t or 36%. (In Scenario I, emissions rise over the same period by 82 t or 21%.)

That PFC emissions do not rise but even drop distinctly in absolute terms is due mainly to developments in the semiconductor industry. This emits in the year 2010 only 11 t CF_4 and 11 t C_2F_6 (Table 65, lines 2 and 6). Compared to Scenario I, this is 66 t less CF_4 and even 90 t less C_2F_6 . In the reduction Scenario II, the emissions of these two PFCs together are slightly lower in the year 2010 than in the baseline year 1995. C_3F_8 emissions from semiconductor manufacturing remain at low levels throughout (Table 65, line 7).

In aluminium smelting, the difference between the reduction scenario and the businessas-usual scenario is less marked than in semiconductor manufacturing. Owing to conversion from side feeding to centre point feeding technology, CF_4 emissions drop in Scenario II by 85 t (cf. Table 65, line 1), compared to a drop of 67 t in Scenario I. In conjunction with this, C_2F_6 emissions from aluminium electrolysis drop in the reduction scenario from 21 t to 12 t (Table 65, line 5), instead of to 14 t.

Table 66: 1995-2010 PFC emissions by source					
in reduction Scenario II in million t CO ₂ equivalent					
Source	1995	2000	2005	2010	
1. Aluminium smelters	1.551	1.165	0.969	0.922	
2. Semiconductor man.	0.174	0.468	0.446	0.193	
3. Printed board man.	0.022	0.022	0.022	0.022	
4. Refrigerant blends	0.015	0.103	0	0	
Total	3.757	3.758	3.442	3.147	

It follows from the assumptions of Scenario II that PFC emissions from printed circuit board manufacture and refrigeration do not develop differently than in Scenario I.

The global warming contribution of PFC emissions is considerably reduced in the reduction scenario as compared to the business-as-usual scenario. By 2010, emissions do not rise as in the business-as-usual scenario to 2.52 million t CO_2 equivalent, but drop to 1.137 million t. Compared to the 1995 baseline (1.762 million t CO_2 equivalent) this is a reduction by 0.62 million t CO_2 equivalent or 35%.

Diagram 33 illustrates the difference between Scenarios I and II.



Diagram 33: Comparison between Scenarios I and II: 1995-2010 PFC emissions in million t CO_2 equivalent. Above all due to the far-reaching substitution of PFC usage in semiconductor manufacturing, emissions in the reduction scenario (Scenario II) drop from 2000 onwards, while in business-as-usual Scenario I emissions rise to 2.5 million t CO_2 equivalent in the year 2010.



Diagrams 34 and 35 illustrate the major role of the semiconductor manufacturing industry in emissions reduction in reduction Scenario II as compared to business-as-usual Scenario I.

Diagram 34: 1995-2010 PFC emissions by source in business-as-usual Scenario I in tonnes per year. The drop in emissions from aluminium electrolysis is over-compensated by the strong rise in emissions from the semiconductor industry (dark area).



Diagram 35: 1995-2010 PFC emissions by source in reduction Scenario II in tonnes per year. The reduction potential (white area above) is due mainly to falling PFC emissions from semiconductor manufacturing, which since the year 2000 no longer rise.

Annex: The years 1995 to 1997

Tables 67 and 68 serve to complete the environmentally relevant emission data for the years 1996 and 1997.

1995-1997 emissions in t per annum

Table 67: 1995-1997 sectoral PFC emissions in t/a					
1995 1996 1997					
1. Aluminium smelting*	230	218	160		
2. Semiconductor man.**	22	39	50		
3. Printed circuit board man. (CF ₄)	0	0	0.1		
4. Refrigerant blends (C_3F_8)	2	5	8		
Total	258	265	221		

* Total of CF_4 and C_2F_6 . ** Total of CF_4 , C_2F_6 and C_3F_8 .

1995-1997 global warming contribution

Table 68: 1995-1997 sectoral PFC emissions in million t CO ₂ equivalent					
	1995	1996	1997		
1. Aluminium smelting*	1.551	1.471	1.076		
2. Semiconductor man **	0.174	0.307	0.399		
3. Printed circuit board man. (CF ₄)	0.022	0.022	0.022		
4. Refrigerant blends (C_3F_8)	0.015	0.032	0.054		
Total	1.762	3.8282	3.548		

* Total of CF_4 and C_2F_6 . ** Total of CF_4 , C_2F_6 and C_3F_8 .

1 CF₄ and C₂F₆ from Aluminium Electrolysis

Aluminium smelting is the main source of CF_4 emissions, and also generates a considerable part of C_2F_6 emissions. These emissions are not a consequence of any targeted use of the gases in the production process, but arise as a byproduct of the electrolytic reduction of alumina (aluminium oxide) to aluminium.

1 The Anode Effect

During the process of aluminium electrolysis as such, the combination of carbon with fluorine to form CF_4 and C_2F_6 does not occur. The electrolyte does indeed contain fluorine, but this does not participate in the reaction at the carbon electrodes. Only under certain conditions do organic fluorides form, albeit at regular intervals of hours or several days. These conditions are the anode effect.

1.1 Aluminium Electrolysis

The solvent of aluminium electrolysis is a mixture consisting mainly of molten cryolite (Na_3AIF_6) . This ensures that the aluminium oxide dissolved in it does not melt only beyond temperatures of 2050 °C, but already below 970 °C.

A typical electrolysis cell is contained in a steel shell that is up to 12 metres long, 4 metres broad and 1-1.2 metres deep (Ullmann 1985, 466). Its bottom is connected as a cathode and lined with carbon, because other materials would be destroyed rapidly by the hot fluoride melt. The anode is positioned above the cell, and consists of 14 to 24 large carbon blocks fitted to two rows of current-supplying rods and immersed in the approx. 20 cm deep electrolyte bath. In addition to cryolite and further fluoride additives (in order to reduce melting temperature), the electrolyte contains alumina (aluminium oxide) at concentrations ranging between 1.5 and 5 percent by weight. The flow of current from the anode brings about the electrolysis of the alumina, namely the deposition of aluminium at the cathode and of oxygen at the anode. The oxygen is discharged as a gas and reacts (burns) with the anode carbon to form CO_2 . The liquid metal collects at the bottom in a metal bath of up to 20 cm height, from which it is regularly extracted.

A firm <u>crust</u> of "frozen" electrolyte forms at the top of the bath and around the anode blocks. This layer, which is up to 10 cm thick, protects the bath from heat losses and vapour emissions, and protects that part of the red hot anode which is not immersed in the bath from oxidation with the air. The disadvantage of the crust is that each time alumina is fed to the cell, the crust needs to be broken open mechanically, which leads to fluctuations in the bath concentration because parts of the crust fall in. The solid crust fragments dissolve slower in the bath than the alumina feedstock powder. They can collect as a hard sludge layer at the bottom of the metal bath, impeding the passage of the current and thus compromising aluminium yield.

In an aluminium smelter, 150 to 600 electrolysis cells (or "pots") are usually positioned in several long rows called cell lines. These cells are supplied with a constant current of 100 to 200 kA. The direct current between the electrodes has a voltage of 4-5 V (Grjotheim/Kvande 1993).

Alumina is fed to the bath in step with its consumption. However, exact dosage is difficult. Excess alumina concentration increases the danger of sludge formation through undissolved quantities. Too low concentration can trigger the "anode effect". An anode effect occurs in aluminium electrolysis when the alumina content in the electrolyte falls below a minimum concentration of approx. 1.5% (Grjotheim/Kvande 1993, 210).

The anode effect is characterized by a strong rise in voltage. The gas bubbles below the anode blocks expand, with the result that the immersed part of the carbon blocks is no longer covered by the bath but by an insulating gas layer. This increases the electric resistance at the anode/bath interface, thus causing a rise in voltage due to the constancy of amperage (U = I x R). The 5 V normal voltage becomes 30-40 V overvoltage (Tabereaux 1996). This suffices to release fluorine from the electrolyte, which reacts with the carbon of the anode blocks to the gases CF₄ and C₂F₆. The gas at the anode changes its composition from CO₂ to CO with a 5-20% CF₄ proportion (Grjotheim/Kvande 1993, 210). Furthermore, C₂F₆ is also formed at a quantity of about 10% of that of CF₄ (Gibbs/Jacobs 1996, 34).²⁶ The anode effect with the associated CF₄ formation persists as long as the anode gas remains as a film below the anode.

1.3 Quenching and Avoiding the Anode Effect

Immediate addition and fine dispersal of alumina in the electrolyte is only the first step for terminating an anode effect. For electrolysis to continue, the gas layer needs to be removed. This is done in modern cells automatically through upwards, downwards and angular movements of the anode blocks. The anode effect duration is generally two to four minutes. Today, manual intervention, which used to be the rule, is only necessary exceptionally. Then a rod made of fresh wood is immersed in the bath, which generates such strong turbulence that the gas layer is dispersed (Aluminium Rheinfelden 1996). Alternatively, compressed air is blown from steel lances underneath the anode blocks.

The times are past in which the anode effect was the main signal for adding alumina to the cell. Nonetheless, it is welcome in certain cases, for it is capable of dissolving the bottom sludge formed through over-feeding, smoothing the anode surface or burning the coal foam in the bath (coal dust fallen from the anodes) (Barber 1996, 2).

However, today the anode effect is primarily viewed as a disturbance, as it is associated with reduced yield, with temperature rise in the bath and with production interruptions. It has become possible to greatly reduce it since - from 1970 onwards - new technologies have become available for continuous and microprocessor controlled alumina feeding. This prevents sludge-forming over-feeding that would make an anode effect necessary as a cleaning mechanism. Anode effect reduction was further promoted by the switch during the same period from the self-baking Söderberg anodes (hardening in process) by prebaked anodes (hardening prior to process), which limit coal dust arisings.

 $^{^{26}}$ CF₄ is formed throughout the 2–4 minute duration of an anode effect. In contrast, C₂F₆ is only formed during the first minute, and up to a maximum concentration of 1–2% of the gas layer (Tabereaux 1996).

The aim of computer-controlled <u>point feeding</u> (see below) is to avoid as far as possible both the upper alumina concentration limit with the danger of sludge formation, and the lower limit with the danger of an anode effect. While this cannot reduce the duration of anode effects, it can reduce their frequency.

1.4 Alumina Feeding - The Key to Reducing Anode Effect Frequency

Since the last cells with self-baking Söderberg anodes in Germany were decommissioned in 1990, only three cell types remain that differ with regard to their anode effect frequency. All use prebaked anodes. The differences of relevance to the anode effect lie in the manner of alumina feeding, which can be from the side, through the centre or through point feeding.²⁷

1.4.1 Alumina feeding from the side (SWPB)

The side feeding technology was introduced in the 1960s. In Germany, 70,000 t primary aluminium production capacity continue to use this technology. It is called "Sidework Prebake" (SWPB), alumina feeding being from the side and the anodes prebaked. After opening the cell covers, a vehicle moves along the cell and breaks open the crust. Alumina feeding is at intervals of 4 to 8 hours and is accordingly rough, which promotes sludge formation. Anode effect frequency is about 1 per cell-day (Aluminium Rheinfelden 1996). This results in typical PFC emissions for the SWPB technology of 1-1.5 kg CF₄ per t primary aluminium (IPAI 1996). In Germany, however, aluminium producers claim that this value is distinctly reduced.²⁸

1.4.2 Simple feeding through the centre (CWPB)

Centre feeding, a technology also developed in the 1960s, is now used since 1990 by all other cells in Germany. Here a distinction can be made between the original centre feeding technology, "Centrework Prebake" (CWPB), and its refinement to centre point feeding ("Point Feeding Prebake", PFPB). With the original CWPB technology, a support structure running above the centre of the cell bears mechanical feeders. Bars or picks break the crust by thrusting down between the two anode block rows. The alumina is fed in measured portions from above through the opening in the crust. Feeding intervals are shorter than in SWPB, at about 60 minutes. Anode effect frequency is between 0.5 and 1 per cell-day (VAW 1996). This results in typical PFC emissions for the CWPB technology of 0.350-0.600 kg CF₄ and a further 0.035-0.060 kg C₂F₆ per t primary aluminium.

1.4.3 Centre Point Feeding (PFPB)

Point feeding ("Point Feeder Prebake", PFPB) is a refinement of CWPB that was introduced in the 1970s (Grjotheim/Kvande 1993, 80/81). Perpendicular picks positioned between the anode blocks which break the crust at several points simultaneously and feed alumina are operated almost continuously at intervals of only a few minutes and with alumina charges of only 0.2 to 2 kg (ibid., 81). Owing to the

²⁷ The directory "Primary Aluminium Smelters and Producers of the World", which is updated several times each year, gives data on the feeding technologies used in individual smelters.

²⁸ According to the German Federal Environmental Agency (Umweltbundesamt, 1999), the emission factor in 1998 in German smelters was only 0.51 kg C_{F_4} and 0.051 kg C_2F_6 per t primary aluminium.

brief intervals, the feeding hole scarcely crusts over, which helps to prevent imprecise alumina feeding through crust entry. Computer control permits feeding according to requirements and a relatively constant alumina concentration in the bath, so that anode effects for bath control are largely dispensed with. In cells using point feeder technology, anode effects only occur 1 to 3 times weekly, or 0.15 to 0.4 times per cell-day (Aluminium Essen 1996). This results in typical PFC emissions for point feeder cells of 0.1 to 0.25 kg CF₄ and an additional 0.01-0.025 kg C₂F₆ per t primary aluminium.

2 1995-1997 Emissions in Germany

Over the 1995-1997 period, the overall capacity of the five German aluminium smelters remained constant at about 580,000 t/a. Primary aluminium production was close to maximum capacity in these three years (see Table 69, last line). Nonetheless, CF_4 emissions dropped over the same period from 209 to 145 t (see Table 69). C_2F_6 emissions dropped accordingly from 21 to 15 t. The global warming contribution dropped over the 1995-1997 period from 1.552 to 1.076 million t CO_2 equivalent.

Table 69: 1995-1997 PFC emissions from aluminium smelters in Germany				
	1995	1996	1997	
CF ₄ [t/a]	209	198	145	
C ₂ F ₆ [t/a] *	21	20	14,5	
Emiss. in mill. t CO ₂ equiv.	1.552	1.471	1.076	
Aluminium production [t]	575,000	576,500	571,900	

Sources: For CF_4 Umweltbundesamt 1999. For aluminium production GDA 1999.

* C_2F_6 was not measured, but calculated as 10% of CF_4 emissions.

The drop in emissions is attributable to the modernization of cells in German smelters, which progressed rapidly since late 1994. The main aim of this was to switch from centre feeding (CWPB) to the more energy efficient and less anode effect generating point feeding (PFPB) technology. In 1995 only a third of the CWPB cells were equipped with point feeders, in 1996 it was already half, and by 1997 conversion was completed (GDA 1999). Of the overall smelter capacity of 580,000 t/a, 510,000 t had thus been modernized to state-of-the-art alumina feeding technology. The remaining 70,000 t/a capacity are cells with side feeding (SWPB), for which retrofitting to point feeding would require complete redesign. Such retrofitting is not anticipated for the foreseeable future.

Empirical survey of CF₄ emissions in 1996

In the second half of 1996, the member companies of the primary aluminium chapter in the federation of German aluminium industries (Fachverband Primäraluminium im Gesamtverband der Deutschen Aluminiumindustrie, GDA) carried out CF_4 emissions measurements in all German production facilities. CF_4 emissions were measured directly during individual anode effects. On this basis, typical CF_4 emissions per anode effect were determined for specific types of operation. As anode effects have been recorded in all companies for many years, annual CF_4 emissions per company could then be determined as the product of the number of anode effects and the company-specific CF_4 quantity per anode effect - and not only for the year of measurement, but for all previous years in which anode effects had been statistically recorded.

Since a large-scale empirical survey of CF_4 emissions carried out in the year 1996 (see box), technology-specific emission coefficients are available for German smelters for the three technologies SWPB, CWPB and PFPB (cf. Table 70).

Table 70: Alumina feeding technology specific CF₄ emission coefficients in aluminium smelters in Germany [kg CF₄ / t Al]				
Alumina feeding technology	SWPB	CWPB	PFPB	
kg CF ₄ / t aluminium	0.51	0.40	0.22	

Source: Umweltbundesamt 1999.

Abbreviations: SWPB: Sidework Prebake; CWPB: Centrework Prebake; PFPB: Point Feeder Prebake. Prebake is in contrast to electrodes which bake during the process.

The CF₄ emission coefficients range from 0.51 kg CF₄ / t Al with SWPB to 0.22 kg CF₄ / t Al with PFPB technology. With these figures and data on the shares of the three technologies in annual aluminium production, CF₄ emissions can be estimated relatively reliably for several years without renewed measurements.

For 1997, we thus arrive for 67,000 t Al from SWPB cells at 34 t CF₄ (67,000 x 0.51 kg) and for 505,000 t Al from PFPB cells at 111 t CF₄ (505,000 x 0.22 kg). CF₄ emissions totalled 145 t. These were joined by a further 10% C_2F_6 emissions.

3 2010 Business-as-Usual Emissions Forecast - Scenario I

The business-as-usual emissions forecast for 2010 assumes that the primary aluminium capacities available in Germany in 1998 remain unaltered in volume and composition. Unaltered composition means that the SWPB cells, with a capacity of 70,000 t/a, are not retrofitted over that period to PFPB technology. The unaltered volume from 1998 onwards requires a brief explanation: The capacity of 1998, amounting to 632,000 t/a (production in the year 1998 figured 612,000 t), is 52,000 t/a higher than that of the 1995-1997 period. It is important to stress that this expansion involved neither the new construction of a smelter nor the restart of decommissioned smelters. What occurred was rather that one of three cell lines which had been shut down several years previously in an existing smelter was returned to production after thorough modernization (Aluminium Essen 1999).

Since the early 1990s, smelters in Germany only cover approx. 50% of German consumption - as compared to 70% in the first half of the 1980s (Metallstatistik 1994, 71). Despite this, it is not to be expected that new smelters are built or decommissioned ones are started up again. If increased demand leads to completely new capacities, then these can be expected to be built, if at all, abroad.²⁹

On the other hand, it does not appear probable at present that over the medium term any capacities will be shut down. While the smelters abroad operate at lower cost, the German smelters retain specific benefits owing to their integrated operations. They have integrated foundries and produce not only simple aluminium ingots, but are also

²⁹ The last new construction of an electrolysis smelter in Germany was in 1973. Since then, new construction with German participation has been carried out exclusively abroad. Between 1989 and 1992, one smelter with 215,000 t/a capacity was erected in Canada, and between 1991 and 1993 an Australian smelter was expanded by 120,000 t/a capacity (VAW-GB 1989, 31; 1990, 22; 1993, 28).

able to supply alloys and intermediate products tailored to customer demands (Aluminium Essen 1996).

Now that since 1997 all cells in German smelters, except for 70,000 t/a SWPB capacity, are equipped with PFPB technology, no comparable modernization leaps are to be expected over the medium term that might lead to significant reduction of anode effects and of the associated PFC emissions. Instead, continuous, minor improvements to the existing technologies (improved cell control etc.) can be expected.

The quantitative assumptions for the business-as-usual Scenario I are as follows:

- 1. The production capacity of aluminium smelters in Germany remains at 632,000 t/a. This capacity is composed of 70,000 t/a SWPB and 562,000 t/a PFPB.
- 2. On the basis of the experience made in recent years, capacity utilization is assumed to be 98%. Production fluctuations are not considered.
- 3. In the year 1998, the kg CF₄ per tonne aluminium emission coefficient is 0.51 for SWPB and 0.22 for PFPB technology. C_2F_6 is emitted at a rate of 10% of CF₄ emissions. (No further PFC gases are taken into consideration.)
- 4. Continuous refinement of the two feeding technologies leads to an annual reduction of the emission coefficients by 1% compared to each previous year. In 2010, the coefficient for SWPB is 0.46 and for PFPB 0.20 kg CF₄ per tonne aluminium.

Under these conditions, the values shown in Table 71 result for the PFC emissions from 1995 to 2010. It should be kept in mind that from 2000 onwards these values are estimates.

Table 71: 1995-2010 CF ₄ and C_2F_6 emissions from aluminium smelting						
in business-as-usual Scenario I						
1995 2000 2005 2010						
CF ₄ [t]	209	157	149	142		
$C_2F_6[t]$	21	16	15	14		
Million t CO ₂ equiv.*	1.551	1.165	1.108	1.054		

* GWP of CF₄: 6500; GWP of C₂F₆: 9200.

Between 1995 and 2010, CF_4 emissions from aluminium smelting in Germany drop by 32%, namely from 209 to 142 t. C_2F_6 emissions drop correspondingly from 21 to 14 t.

The development of emissions over the 1995-1997 phase and the subsequent period is characterized by a variety of factors. The strong drop between 1995 and 1997 was due to the completion of conversion from simple centre feeding cell technology (CWPB) to modern point feeding (PFPB), the latter generating per tonne aluminium only about half of former CF_4 emissions.

Over the 13 years from 1997 to 2010, PFC emissions drop in total by approx. 10%. The 52,000 t/a growth in capacity which came on line in 1998, and led to more than 11 t of new annual CF_4 emissions, only halted the declining PFC emissions trend briefly. The drop since 1998 is based exclusively upon continuous improvements to the two

alumina feeding technologies, which yield an annual drop in PFC emissions of 1% compared to each previous year. The average emission factor (weighted mean of the two feeding technologies) drops from 1998 to 2010 from 0.257 to 0.229 kg CF₄ / t Al. In 1995, it still figured 0.363 kg CF₄ / t Al.

The global warming contribution of PFC from aluminium smelting drops from 1995 to 2010 from 1.551 to 1.054 million t CO_2 equivalent, being a drop of 32%.

4 2010 Emission Reductions until - Scenario II

The conditions for greater emission reductions than in business-as-usual Scenario I can be stated quite briefly: The reduction scenario is distinguished from the business-as-usual scenario solely by the assumption that the remaining SWPB cells are converted to PFPB technology. It is assumed that this measure is implemented after the year 2000. Scenario II assumes (arbitrarily) the 2003-2005 period for technology conversion.

The assumptions concerning overall capacity, capacity utilization, emission coefficients (kg PFC per t aluminium) and their annual reduction (by 1% compared to each previous year) are the same as in Scenario I.

Retrofitting the remaining 70,000 t production capacity from SWPB to PFPB technology is two and a half times as expensive per cell as retrofitting from CWPB technology (Harnisch et al. 1998, 13). This, however, is only a fraction of what the conversion of Söderberg cells to point feeding cost in the 1980s in Germany. According to the same source, these conversion costs are about ten times as high.

Scenario II does not include any further factors that might reduce PFC emissions. The CF₄ emission coefficient of 0.22 kg CF₄ / t Al taken for point feeding in German smelters in 1998 is not first class when compared to figures found in other countries. The literature reports specific emission factors with best available technology of 0.05 kg CF₄ / t Al and less (inter alia in Harnisch et al. 1998). However, the authors of the present study do not have enough information on the conditions under which such emission factors could be transferred to the whole capacity in Germany. Scenario II also does not consider completely new smelting technologies such as 'inert anodes' made of metal (Beck 1995), which do not generate any PFC emissions.

The assumed retrofitting of the remaining 70,000 t/a capacity from SWPB to PFPB technology results in lower values for PFC emissions than in Scenario I from 2005 onwards (see Table 72).

Table 72: 1995-2010 CF₄ and C₂F ₆ emissions from aluminium smelting				
in reduction Scenario II				
	1995	2000	2005	2010
CF ₄ [t]	209	157	131	124
$C_{2}F_{6}[t]$	21	16	13	12
Million t CO ₂ equiv.*	1.551	1.165	0.969	0.922

* GWP of CF₄: 6500; GWP of C₂F₆: 9200.

In Scenario II, CF₄ emissions drop over the 1995-2010 period from 209 to 124 t. C_2F_6 emissions drop from 21 to 12 t. This is a relative drop of 40% compared to 1995. (In Scenario I, the drop amounts to 32%.)

The global warming contribution drops from 1.551 to 0.922 million t CO₂ equivalent.

Until the year 2000, emissions do not differ from those in business-as-usual Scenario I (cf. Table 71). Scenario II assumes that the emissions-reducing retrofitting from SWPB to PFPB technology only takes place between 2003 and 2005. Consequently, emissions in Scenario II are only below those of Scenario I in the year 2005 and later (cf. Table 72).

The difference between Scenarios I and II in terms of PFC emissions figures 12.5% in the year 2010. This reduction amounts to 18 t CF_4 and 2 t C_2F_6 or 0.132 million t CO_2 equivalent. That the reduction is not greater has two reasons. Firstly, the retrofitting of the 70,000 t/a SWPB capacity to PFPB technology only concerns 11% of overall capacity. Secondly, the difference between the emission factors of the SWPB and PFPB technologies is not very large in Germany (in contrast to other countries). In 1998, the SWPB emission factor figured 0.51, that of PFPB 0.22 kg CF_4 / t Al.

Diagram 36 illustrates graphically from when onwards and to what extent PFC emissions differ between the two scenarios. This shows that the effective reduction in emissions mainly took place between 1995 and 1997 and that the - not very large - reduction potential only becomes effective during the 2000-2005 period.



Diagram 36: 1995-2010 CF₄ and C₂F₆ emissions from aluminium smelters in Germany in t/a, in Scenarios I and II. Between 1995 and 1997, emissions of CF₄ (black lower area) and of C₂F₆ (grey upper area) drop substantially owing to point feeding technology. The slight rise in 1998 is due to capacity expansion. The potential emission reduction in Scenario II (white area) resulting from retrofitting the entire capacity to point feeding becomes effective from 2003 onwards.

2 PFCs as Semiconductor Etching Gases

A discussion of PFC gases and the conditions determining their emissions in the semiconductor industry is given in the final Part IV of this report, where all fluorinated greenhouse gases emitted by the semiconductor industry are treated together. We therefore do not discuss the technologies involved here in detail, but merely present the emissions in brief. The data contained in Tables 73, 74 and 75 are taken from Part IV.

PFCs are used in relatively large quantities as etching gases in semiconductor manufacturing. They are applied mainly in the regular cleaning of CVD (chemical vapour deposition) plasma chambers. The most important PFCs are C_2F_6 and CF_4 . C_3F_8 is also in use since 1997 as a substitute for C_2F_6 . In transformation in the plasma, both C_2F_6 and C_3F_8 lead to the new generation of considerable CF_4 quantities as a byproduct.

The high emissions of C_2F_6 are remarkable - in 1996 these already exceeded the quantities emitted from aluminium electrolysis in Germany (cf. Table 73, line 2). In the business-as-usual scenario (Table 74) they rise to 101 t by the year 2010. The rise of CF₄ emissions to 77 t in the year 2010 in the business-as-usual scenario is also considerable. Under the assumptions of the business-as-usual scenarios, PFC emissions (CF₄, C₂F₆ and C₃F₈) from semiconductor manufacturing will exceed those from aluminium smelting (cf. Table 71) in the year 2010 by almost 0.4 million t CO₂ equivalent.

The comparison of emissions in 2010 in the two respective reduction scenarios presents a different picture. Here PFC emissions from semiconductor manufacturing only figure 0.193 million t CO_2 equivalent (Table 75), while PFC emissions from aluminium electrolysis are 4.5 times as large: 0.922 million t CO_2 equivalent (Table 72).

Table 73: 1995-1997 PFC consumption and emissions in semiconductor manufacturing						
	1995 1996 1997					
	Consump.	Emission	Consump.	Emission	Consump.	Emission
1. CF ₄ [t] *	15	12	29	21	34	23
2. C ₂ F ₆ [t]	20	11	37	19	56	27
3. C₃F ₈ [t]	0	0	0	0	0.3	0.1
Total	35	23	66	40	93	51
Mill. t CO2 equiv.		0.174		0.307		0.399

1 1995-1997 Consumption and Emissions in Germany

* Untransformed process gas plus new formation by plasma decomposition of C_2F_6 and C_3F_8 .

The difference between consumption and emissions results firstly from the only partial chemical transformation in the plasma reactor, and secondly from the effect of downstream exhaust gas treatment.

2 2010 Business-as-Usual Emissions Forecast - Scenario I

The assumptions for Scenarios I and II are also presented in Part IV. Here only the resultant emissions are presented.

In business-as-usual Scenario I, the emissions shown in Table 74 result until 2010:

Table 74: 1995-2010 PFC emissions from semiconductor manufacturing				
	1005	2000	2005	2010
	1990	2000	2003	2010
	12	31	50	11
2. $C_2 F_6 [t]$	11	36	62	101
3. C ₃ F ₈ [t]	0	1	2	3
Total	23	68	114	181
Million t CO2 equivalent	0.174	0.542	0.904	1.445

* Untransformed process gas plus new formation by plasma decomposition of C_2F_6 and C_3F_8 .

Over the 1995-2010 period, PFC emissions rise from 23 to 181 t.

3 2010 Relative Emission Reductions - Scenario II

In the reduction scenario, the lower emissions shown in Table 75 result from 2000 onwards. They total for all PFCs in the year 2010 25 t PFC or 0.193 million t CO_2 equivalent.

Table 75: 1995-2010 PFC emissions from semiconductor manufacturing in reduction Scenario II				
	1995	2000	2005	2010
1. CF ₄ [t] *	12	27	27	11
2. C_2F_6 [t]	11	31	28	11
3. C ₃ F ₈ [t]	0	1	2	3
Total	23	59	57	25
Million t CO ₂ equivalent	0.174	0.468	0.446	0.193

* Untransformed process gas plus new formation by plasma decomposition of C_2F_6 and C_3F_8 .

The reduction scenario leads by the year 2010 to PFC emissions that are reduced by 156 t or 86% compared to Scenario I.

The global warming contribution then figures only 0.193 million t CO_2 equivalent instead of 1.445 million t (Scenario I).

The main cause of the - feasible - emissions reduction is the replacement of PFCs by NF_3 in chamber cleaning. NF_3 , for its part, can, in contrast to PFCs, be dissociated to a rate of 98-99% through highly effective and economical new plasma technologies, without recombination (Air Products 1999).

It is to be noted that the additional NF_3 emissions which result in the reduction scenario as a consequence of PFC phase-out only figure 1 t in the year 2005 and about 2 t in the year 2010.



Diagram 37: 1995-2010 PFC emissions from semiconductor manufacturing: Comparison between Scenarios I and II. In contrast to the business-as-usual Scenario I, in which emissions rise to 181 t (2010), emissions remain constant in the reduction scenario from 2000 onwards and drop from 2009 onwards. The difference results by the year 2010 in a reduction potential (white area) of 156 t.



Diagram 38: 1995-2010 PFC emissions from semiconductor manufacturing in million t CO_2 equivalent: Comparison between Scenarios I and II. In the business-as-usual scenario, emissions rise to 1.445 million t CO_2 equivalent by 2010, while in the reduction scenario they only amount to 0.193 million t in that year.

3 CF₄ in Printed Circuit Board Manufacturing

CF₄ has a further intentional use in electronics as a plasma etching gas, namely for desmearing in printed circuit board manufacturing.

Desmearing means cleaning drilled holes in multilayer printed circuit boards in lowpressure plasma chambers. The plastic slivers of epoxy or polyimide resins in the boards, which consist of alternate layers of plastic and copper, are etched away in order to allow improved connection of the copper in the subsequent wiring (plasonic 1999).

This process is applied instead of wet-chemical processes by larger printed circuit board manufacturers. The plasma gas generally consists of 20-30% CF₄ and 70-80% oxygen (Technics Plasma 1996). As yet, there are only few plants with continuous round-the-clock operation. Most plants are operated discontinuously (batch operation) with gas throughputs of approx. 100 ml per minute. Their number in Germany is estimated at about 50 (Buck Plasma Electronic 1996).

Experts estimate the annual CF₄ consumption in Germany for this process at 4 tonnes in the year 1995.³⁰ No growth or decline is anticipated for the future (plasonic 1999).

In the printed circuit board manufacturing industry, there are alkaline exhaust gas scrubbers for capturing the reaction products, but no systems that could destroy unutilized or recombined CF_4 in the exhaust gas (plasonic 1999). If the same utilization removal efficiency is assumed for CF_4 in the plasma reactor as in semiconductor manufacturing, namely 15% (Air Products 1999), then 85% of the CF_4 gas supplied is emitted unchanged or recombined in the exhaust gas from the process chamber.

Table 76: 1995-2010 CF₄ consumption and emissions in printed circuit board manufacturing					
1995 ff. 2010					
Consumption [t] 4 4					
Emissions [t] 3.4 3.4					
Emissions in million t CO_2 equiv.0.0220.022					

Sources: see text

Of the 4 t CF₄ consumed annually, 3.4 t is emitted, being 0.022 million t CO₂ equivalent.

These emissions are expected to be constant over the medium term (until 2010), so that no separate estimate of emissions in a business-as-usual and a reduction scenario is carried out here.

³⁰ CF₄ is also used in small quantities for plasma cleaning other than in electronics, namely as an additive to oxygen for degreasing metal surfaces.

4 C₃F₈ as Refrigerant

 C_3F_8 (R-218) is in use since 1993 as a blend component in drop-in refrigerants for CFC substitution. Drop-in means that the existing refrigeration or air-conditioning equipment which previously operated on CFCs can continue to operate with the PFC-containing blend without major equipment modification.

Although several producers (DuPont, ICI, Elf Atochem) offer refrigerants containing the PFCs R-116 and R-218³¹, only those of the producer Rhodia are sold in Germany in anything more than negligible amounts. R-218 is used in proportions between 9% and 39% in three Rhodia blends, the main components being in two cases HFCs³² and in one case an HCFC:

- In the R-12 substitute R-413A, with $9\% C_3F_8$ in addition to R-134a and isobutane;
- In the R-502 substitute R-403B, with 39% in addition to R-22 (HCFC!) and propane;
- In the R-13B1 substitute "Isceon 89", with 9% in addition to R-125 and propane.

The main field of application of R-413A is R-12 substitution in stationary commercial refrigeration and mobile air conditioning (Schwarz/Leisewitz 1998). Isceon 89 is used as a drop-in for deep-freezing equipment. The blend R-403B is used as a substitute for the CFC-containing refrigerant R-502.

As the bulk of R-218-containing refrigerants used in Germany comes from only a single supplier (Rhodia), R-218 consumption and emission figures are not treated separately for the three refrigerant blends, but are presented in aggregate form.

1 1995-1997 Consumption and Emissions in Germany

Table 77: 1995-1997 C₃F ₈ (R-218) consumption and emissions as refrigerant blend component						
	1995		1996		1997	
	Consump.	Emission	Consump.	Emission	Consump.	Emission
C ₃ F ₈ (R-218) [t]	11	2	17	5	20	8
Mill. t CO ₂ equiv.		0.015		0.032		0.054

Consumption of R-218 as a blend component rose over the 1995-1997 period from 11 to 20 t. Emissions, whose emission rates for operating and disposal emissions are the same as in Section 1 of Part I of the present report, rose over the same period from 2 to 8 t. Their global warming contribution rose accordingly from 0.015 to 0.054 million t CO_2 equivalent.

³¹ These are the R-22-containing refrigerant blends 412A and 509A, and the R-23-containing 508A and 508B. R-22 is an ozone-depleting substance. R2-3 has a very high GWP.

³² The HFCs in these blends (R-134a and R-125) are discussed in Part I of the present study.

2 2010 Emissions Forecast

The three C_3F_8 -containing refrigerants are all substitutes for fully halogenated CFCs used in order to continue to operate old equipment. Their new application will only continue as long as the CFC substitution process continues. This has presently, i.e. in 1999, been concluded, with only a few exceptions. New consumption has thus dropped to the level of refills for operating losses. Emissions drop further in the course of the decommissioning of refrigeration and air-conditioning equipment. As this is old equipment, no emissions are to be expected any longer after 2005.

The HCFCs in refrigeration and air-conditioning systems must albeit continue to be substituted, notably HCFC-22. This is already banned in new equipment from 2000 onwards, and must also be substituted by zero-ODP refrigerants in old equipment by the year 2010. However, the substitute for R-22, which Rhodia offers under the name Isceon 59, is a blend of HFC-125, 134a and isobutane, and thus contains no PFC (Rhodia 1998). The substitution of HCFCs is thus not expected to lead to any new consumption and emissions of R-218, unless other suppliers enter the market with noteworthy quantities of PFC-containing blends. This possibility is excluded in the following.

As shown in Table 78, PFC (R-218) emissions still figure 15 t in the year 2000. By the year 2005, they drop to 0.03 t. From then on, emissions are zero.

Table 78: 1995-2010 $C_{3}F_{8}$ (R-218) emissions as refrigerant blend component					
in business-as-usual Scenario I					
	1995	2000	2005	2010	
C ₃ F ₈ (R-218) [t]	2	15	0.03	0	
Ems. in mill. t CO ₂ equiv.	0.015	0.103	0.000	0	

In the year 2000, when emissions peak, their global warming contribution figures 0.103 million t CO_2 equivalent.

A distinction according to two scenarios as carried out for HFC emissions from refrigeration and stationary air conditioning (business-as-usual on the one hand and emission reduction by step-wise introduction from 2002 onwards of mandatory maintenance for refrigeration and air-conditioning equipment on the other) is practically irrelevant for R-218-containing refrigerant blends, because their emissions tend towards zero in any case from 2002 onwards.

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IV. Fluorinated Gases in Semiconductor Manufacturing

Preliminary note

Of the principal fluorinated gases used in semiconductor manufacturing, NF₃ is discussed here for the first time. The other gases have already been discussed in previous sections: CF_4 , C_2F_6 in aluminium electrolysis, CHF_3 and C_3F_8 in refrigerants and SF_6 in a variety of applications. The technology-specific treatment of all of these gases in the present Part of this report does not supply any new figures above and beyond those contained in the previous Parts I to III. The function of Part IV is rather to present the basis for the findings with regard to consumption and emissions of fluorinated gases in semiconductor manufacturing that were previously presented in summary form in Parts I to III. The various fluorinated gases are referred to collectively here - following IPCC 99 - as "fluorinated compounds" (FCs).

Plasma Etching with Fluorinated Compounds

The gases CHF₃, CF₄, C₂F₆, SF₆, NF₃ and - as yet in only small amounts - C₃F₈ are applied either for wafer etching or for chamber cleaning.³³ In wafer etching, fine structures are produced on the surfaces of wafers: microscopic pathways of sub-micron breadth (<0.5 μ m) on thin layers of silicon oxide, silicon nitride or tungsten. In chamber cleaning, silicon- and tungsten-containing layers are removed from the walls of reaction chambers: in the chemical vapour deposition (CVD) process, chemicals are deposited on the wafer, but partly also on the chamber walls, from which they need to be removed.

Both wafer etching and chamber cleaning are carried out in low-pressure reaction chambers in which the FCs supplied are induced into a plasma state by high-frequency waves. In this process, among other products, highly reactive fluorine radicals (molecular fragments with free bonds) are formed. These react chemically with siliconor tungsten-containing material and convert the solids (e.g. SiO_2) into volatile fluorides (e.g. SiF_4), which are extracted as exhausts from the chamber.

Chemically speaking, both wafer etching and CVD chamber cleaning are etching processes. In practice, however, only wafer etching is termed "etching", and chamber etching is termed "cleaning". The various FCs are exclusively relevant as a source of fluorine, and are in principle exchangeable. However, SF_6 and CHF_3 are better suited for etching, and C_2F_6 , NF_3 and C_3F_8 for cleaning. Only CF_4 is used for both. Plasma chambers are designed for specific etching or cleaning gases through their construction, volume, gas flow, pressure, frequency etc., so that in already installed facilities etching gases are rarely substituted.

1995-1997 Consumption in Germany

Over the 1995-1997 period, FC consumption in Germany for semiconductor manufacturing rose steeply (Electronics Gases Survey). The total grew from 50 to 110 t

³³ Other FCs used in only small amounts for etching, such as C_4F_8 , C_3HF_5 or BF_3 , are not considered further here. Nor is WF₆ (used for metal deposition), which is not a greenhouse gas.

or by 120%. Table 79 shows that two gases played the prime role in this rise: C_2F_6 and CF_4 . C_2F_6 consumption rose disproportionately from 20 to 56 t. C_2F_6 is the most used FC, with a share of 50% in total FC consumption. CF_4 , the second most important FC, accounted for one third of usage in 1997, this also being disproportionate growth from 1995. The other FCs, amounting to 20 t, were much less important in 1997.

Table 79: 1995-1997 consumption of fluorinated etching gases (FCs)						
in semiconductor manufacturing [t/a]						
	GWP	1995	1996	1997		
SF ₆	23900	7	9	10		
CHF ₃	11700	5	6	7		
CF ₄	6500	15	29	34		
C_2F_6	9200	20	37	56		
NF ₃	8000	3	3	3		
C ₃ F ₈	7000	0	0	0.3		
Total		2045	2080	2110		

Source: Electronics Gases Survey

 C_2F_6 is used almost exclusively and CF_4 predominantly for CVD chamber cleaning. The disproportionate growth of both FCs, which compete with NF₃ and C_3F_8 , is due to the miniaturization of semiconductors, which places increased purity requirements upon the process chambers. In the course of the move from 200 mm to 300 mm diameter wafers, which is viewed as a 'technology leap', cleaning intervals will continue to shorten, so that the demand for chamber cleaning gases must be expected to continue to grow disproportionately.

In 1997, some 75% of the FCs consumed in semiconductor manufacturing were used for chamber cleaning, 25% for wafer etching.³⁴ This corresponds to the consumption in Germany of SF_6 , CHF_3 and CF_4 , of which a third is used for wafer etching (Zörmer 1999; Hollenbach 1999).

3 1995-1997 Emissions in Germany

Two factors determine how much of the FC input is released to the atmosphere unaltered and thus contributes to global warming. The first is the utilization removal efficiency (URE) of FC transformation in the plasma chamber. The second is the prevalence and efficiency of downstream exhaust gas treatment systems to destroy that fraction of the FCs which remains untransformed in the plasma.

3.1 FC Utilization Removal Efficiencies in Plasma

Utilization removal efficiency of the individual FCs varies. There are no fixed values, but for each gas there is at least a certain range within which the URE will remain, depending upon the concrete conditions of application. It can generally be stated that URE depends upon molecular stability and - with regard to perfluorocarbons (PFCs) - rises with the number of fluorine atoms. The hierarchy for PFC rates is as follows:

³⁴ These figures correspond to the international experience: "Currently about 60-80% of PFCs consumed in the semiconductor industry are for cleaning process chambers" (Ridgeway et al. 1998).

 $C_3F_8 > C_2F_6 > CF_4$ (Theirich 1996). The URE of CHF₃ (17%) is in the region of that of CF₄ (15%), that of SF₆ (27%) is in the region of that of C₂F₆ (32%). NF₃ and C₃F₈ have the highest URE rates, at 62% and, respectively, 60%. Table 80 lists average URE rates for FCs under typical conditions of application as reported in the literature:

Table 80: Typical FC utilization removal efficiency (URE) rates in plasma						
[% of FC input]						
FC	SF_6	CF_4	CHF ₃	C_2F_6	NF_3	C_3F_8
URE	27%	15%	17%	32%	62%	60%
CF₄ byproduct creation				10%		25%

Sources: Air Products 1996; Zazzera/Reagan 1997; Conners/Raoux 1998.

Low URE rates - such as 15% for CF_4 - mean that the greater part of the etching gas introduced to the chamber is discharged as exhaust. High URE - such as 62% for NF₃ means that the initial gas is largely transformed and only a smaller part still remains in the exhaust. A high proportion of the initial gas in the exhaust need not always mean that little decomposition has taken place in the plasma. This is because in the plasma the molecular fragments partially combine again with each other instead of reacting with the silicon (CF₄ $CF_3 + F$). One reason for the high URE of NF₃ (62%) is that no recombination occurs after its breakdown into nitrogen and fluorine radicals.

Recombination is also the reason why the decomposition products of C_2F_6 and C_3F_8 , although these gases have URE rates of 32% and 60%, partly create an FC again, namely CF₄. Some 10% of C_2F_6 usage is transformed into CF₄, and even 25% of C_3F_8 (see Table 80). C_2F_6 and C_3F_8 thus contribute not only through their undecomposed fractions to global warming, but additionally through CF₄ formed in the process.³⁵

FCs that recombine or are not utilized in the chamber represent <u>potential</u> emissions. To estimate <u>effective</u> emissions, the exhaust gas treatment systems installed to destroy surplus FCs must additionally be taken into consideration.

3.2 FC Waste Gas Treatment

Prior to 1993, waste gas treatment consisted only of scrubbers for <u>reaction products</u>. These largely retain the water-soluble exhausts SiF_4 , SiH_4 , $SiOF_2$, COF_2 , SO_2 etc. in aqueous alkaline solution. However, they let the fluorinated gases not converted in the reactor - which are all water-insoluble - pass unhindered.

Exhaust systems for FCs are as yet almost all based upon high-temperature destruction. The exhaust is not conveyed directly from the vacuum pump to the scrubber, but first to a combustion chamber. The fuel used in the high-oxygen environment is hydrogen or - owing to its hydrogen content - natural gas. At temperatures around 1500 °C, the FCs decompose, and their components combine with H₂ or O₂. Ideally, they are converted completely to HF on the one side and CO₂ (for CF₄, CHF₃, C₂F₆, C₃F₈), SO₂ (for SF₆) or N₂ (for NF₃) on the other. The alkaline scrubber

³⁵ It may be noted that small quantities of further perfluorinated organic byproducts can be created from organic fluorinated etching gases, such as C_3F_6 , C_4F_6 or, under certain conditions, the toxic i- C_4F_8 from CHF₃ (cf. Tiller et al. 1992).

neutralizes HF and the SO₂ formed in SF₆ breakdown. Liquid effluent remains to be disposed of. The exhaust from the combustion chamber contains CO₂, H₂O and nitrogen.

The burner exhaust systems are reported to have high destruction removal efficiency (DRE) for FCs, figuring 98% to 99% under real conditions of operation (Infineon 1999). Only CF_4 is problematic for the exhaust gas systems, due to its high molecular stability, with only 95% DRE (Sematech 1995).

Exhaust gas treatment systems are currently not in such widespread use that all FC chamber emissions might be destroyed. In 1997, the systems (burner systems) were essentially restricted to new fabrication facilities in operation since 1995. In relation to all potential FC emissions, the surveyed manufacturers of exhaust gas systems assume for 1997 a system saturation rate of 30%. In 1995, this rate was 20%, in 1996 25% (Centrotherm 1999; Edwards 1999; DAS 1999, CS 1999).

3.3 Emissions

Effective annual FC emissions can be estimated on the basis of annual consumption figures. This estimation requires two main steps for each FC:

- First, the specific URE rates shown in Table 80 are applied to the annual consumption levels shown in Table 79. This gives the decomposed FCs whose inorganic decomposition and reaction products enter the scrubber. The FCs newly created or undecomposed in the plasma are <u>potential</u> emissions.
- In the second step, the system saturation rates of exhaust gas treatment systems and their efficiencies of 98% or 95% are applied to the potential emissions. The potential emissions lead on the one side to decomposition products for the scrubber and on the other side to the effective annual FC emissions.³⁶ These emissions are listed for the 1995-1997 period for the individual FCs in Table 81.

Table 81: 1995-1997 effective FC emissions from semiconductor						
manufacturing in t						
	1995	1996	1997			
SF ₆	4	5	5			
CHF ₃	3	4	4			
CF ₄ *	12	20	23			
C_2F_6	11	19	27			
NF ₃	0.9	0.9	0.8			
C ₃ F ₈	0	0	0.1			
Total in t	31	49	60			
Mill. t CO ₂ equiv.	0.316	0.473	0.577			

* Untransformed process gas plus new formation by plasma decomposition of C₂F₆ and C₃F₈.

³⁶ The values for <u>individual</u> FCs only apply assuming an <u>even</u> exhaust gas system saturation for all gases in the year under consideration. This assumption holds true in a first approximation.
In total, FC emissions from semiconductor manufacturing rose over the 1995-1997 period from 31 t to 60 t. Due to higher FC exhaust gas treatment system saturation rates (which rose from 1995 to 1997 from 20% to 30%), the growth in emissions is smaller than the growth in consumption (50 t to 110 t).

In terms of individual FCs, the perfluorocarbons (PFCs) C_2F_6 and CF_4 accounted in 1997 for more than 80% of all emissions, with 50 t; C_2F_6 accounted for 45%, CF_4 for 38%. The difference between CF_4 and C_2F_6 is smaller in emissions than it is in consumption. This is due firstly to the lower rate of CF_4 destruction in exhaust treatment systems and, secondly, to the circumstance that secondary CF_4 created in the plasma decomposition of C_2F_6 and C_3F_8 is emitted in addition to the primary CF_4 applied for etching and cleaning.

The global warming contribution of all FCs together figured 0.316 million t CO_2 equivalent in 1995, 0.473 million t in 1996 and 0.577 million t in 1997 (cf. Table 81). In all three years, the main contribution came from C_2F_6 (GWP: 9200).

3.4 Schematic of FC Flows in 1997

The following Figure gives a graphic representation of the paths of total FCs used in 1997 through to effective emission or decomposition product disposal.



Fig. 3: Flowchart of total FC usage in semiconductor manufacturing in Germany in 1997. Of the 110 t FCs (1), 25 t (2) are broken down to non-FCs in plasma chambers and enter the scrubber for inorganic decomposition products (5). The FCs newly created or not transformed in the plasma amount to 85 t and form the potential emissions (3). Of these, 30% enter FC exhaust gas treatment systems (4), with destruction removal efficiency of 95 or 98%: the decomposition products of 24.7 t FCs also enter the scrubber (5). The remaining 60.3 t FCs (6) are released to the atmosphere, this being just under 55% of the initial 110 t FC input.

4 2010 Business-as-Usual Emissions Forecast - Scenario I

In an industry as dynamic as semiconductor manufacturing, any forecast extending beyond the next two or three years is on shaky ground. Forecasts for a period of twelve years are all the more shaky. The following business-as-usual forecast until the year 2010 represents only one of several possible trajectories. It is only valid under the following five, simplified boundary conditions.

- 1. In step with the high rate of growth of semiconductor manufacturing in Germany, total FC usage grows by 10% annually from 110 t in 1997 to 380 t in 2010, i.e. by 245% over that period.
- 2. The two sub-sectors etching and cleaning grow at different rates:
 - For etching, SF₆, CHF₃ and CF₄ (partial) usage grows steadily, but below average, namely by 6% annually from 1998 onwards.
 - For cleaning, C₂F₆, NF₃ and CF₄ (partial) usage rises disproportionately due to increasing purity requirements, namely by 11% annually from 1998 onwards.
- 3. Consumption of C_3F_8 , which was newly introduced in 1997, rises to 14 t by 2010. This quantity is at the cost of C_2F_6 and - partially - CF_4 .³⁷
- 4. The specific FC utilization removal efficiencies (URE) in the plasma of 1995-1997 remain constant until 2010.
- 5. The FC exhaust gas treatment system saturation rate remains in accordance with present investment plans at 30%, i.e. the 1997 rate. This does not, however, mean stagnation of the 1997 stock, but rather an annual growth of newly installed FC exhaust treatment systems by 10%, as FC usage grows annually by 10% (see assumption 1).

Under these assumptions - which, except for differing growth rates of FC usage for etching and cleaning, anticipate largely steady developments of FC usage and FC emissions - the following effective emissions result for the years 1995, 2000, 2005 and 2010 in the business-as-usual Scenario I:

Table 82: 1995-2010 effective FC emissions from semiconductor manufacturing					
in business-as-usual Scenario I					
	1995	2000	2005	2010	
SF ₆	4	6	8	11	
CHF ₃	3	5	6	8	
CF ₄ *	12	37	50	77	
C_2F_6	11	36	62	101	
NF ₃	1	1	2	3	
C ₃ F ₈	0	0.6	2	3	
Total in t	2026	80	130	2213	
Ems. in mill. t CO2 equiv.	0.316	0.756	1.193	1.836	

* Untransformed process gas plus new formation by plasma decomposition of C_2F_6 and C_3F_8 .

 $^{^{37}}$ C₃F₈, which is suitable as a drop-in for existing facilities, permits under certain process conditions a global warming contribution that is reduced by 60% compared to C₂F₆ (SEMATECH 1998; 3M 1999).

Total FC emissions rise to 203 t by 2010. The greater part of this is attributable to the gases C_2F_6 and CF_4 , with 101 t (50%) and 77 t (38%) usage, both of which are used primarily for chamber cleaning. Of the 77 t CF_4 , 15 t come from etching and 62 t from cleaning. Of the latter figure, 51 t are primary gas and 11 t are secondary gas from plasma transformation of C_2F_6 and C_3F_8 .

Of the gases used for etching - SF_6 , CHF_3 and CF_4 (partially) - only 34 t are emitted. In 2010, the ratio of etching to cleaning gases in total effective FC emissions figures about 16% to 84%. In 1995, this ratio was still 35% to 65%.

PFCs (CF₄, C_2F_6 , C_3F_8) have a share in total FC emissions of 181 t in the year 2010, of which 166 t come from chamber cleaning.

The global warming contribution rises to 1.836 million t CO_2 equivalent by the year 2010. Of this, 0.928 million t (50%) come from undecomposed C_2F_6 , and a further 0.066 million t from CF_4 created in C_2F_6 transformation. C_2F_6 thus effectively accounts for a share of almost 1 million t CO_2 equivalent in the global warming contribution of FCs in semiconductor manufacturing.

5 2010 Emission Reductions - Scenario II

Scenario II is guided by the April 1999 decision of the World Semiconductor Council (WSC), the federation of all major semiconductor manufacturers of the world, to cut aggregate absolute FC emissions from semiconductor fabrication globally "by 10% or more by the year 2010". For the European manufacturers the baseline year is 1995 (WSC 1999). Considering the growth in FC emissions in business-as-usual Scenario I from 0.316 to 1.836 million t CO_2 equivalent by the year 2010, this target appears highly ambitious for Germany. It would mean that far from rising, global warming FC emissions would instead have to be returned to approx. 0.3 million t CO_2 equivalent. Nonetheless, in view of a pending technology leap in chamber cleaning and etching gas exhaust treatment, the WSC decision appears by no means illusory. These new conditions are outlined briefly in the following.

5.1 Technology Leaps in FC Usage

 NF_3 is distinguished from the other FCs by the circumstance that its decomposition products created in the plasma do not recombine. The fluorine dissociated from the nitrogen either reacts as intended with the microchip surface layering or is discharged from the chamber as elementary gas (F_2). However, NF_3 is not decomposed completely, but only by about 62%. Untransformed NF_3 is destroyed after the chamber in the exhaust treatment systems. This is done because NF_3 is toxic. The threshold limit value (TLV) applicable for NF_3 in the USA is only 10 ppm.

In contrast to the conventional RF (radio frequency) plasma technology, in a highdensity microwave plasma NF₃ is dissociated 98-99% if it is supplied without admixture of other gases. The basic principle of the new technology based on this is to fit a microwave plasma source at the inlet to the process chamber, which breaks down NF₃ by approx. 98%, so that 98% free fluorine enters the main chamber (Raoux et al. 1999). As it does not recombine there with nitrogen, chamber exhausts consist only of the fluorine-etched deposition residues and elementary nitrogen and fluorine.

With this technology, NF_3 levels in the exhaust are so low that special FC treatment such as combustion - is not always necessary to meet the threshold limit value of 10 ppm (Kopatzki/Guerin 1999). If a dry bed absorber is used, high-temperature combustion is even no longer necessary to treat the aggressive elementary fluorine. These absorbers, containing catalytically active granulates, absorb at room temperature both the fluorine and the fluorine-containing reaction products from CVD cleaning. Such systems are significantly less costly to procure and operate than the combination of burner and scrubber (ibid. 1999).

The leading manufacturers of plasma assisted CVD chamber systems worldwide, Applied Materials and Novellus, have both developed cleaning with upstream dissociated NF₃ to series maturity and are in the process of marketing this technology under the names "Remote Plasma Clean" and, respectively, "Microcleans" (Applied Materials 1999; Novellus 1999).³⁸ As compared to the use of carbon-containing FCs (CF₄, C₂F₆ and C₃F₈), the new process has two advantages. Firstly, the shift of FC decomposition to the beginning of the process, without substantial extra cost, replaces FC exhaust treatment and facilitates the treatment of inorganic exhaust substances. Secondly, cleaning times are up to 60% shorter compared to the C₂F₆ process, and wafer throughput up to 20% higher in the same time (Conners/Raoux 1998).

The increased productivity in semiconductor manufacturing (more wafers in the same time) provided by the new NF₃ technology outweighs several times over the cost drawback of the process gas NF₃, which presently costs four times more per kilogram than C_2F_6 .

In <u>wafer etching</u>, for which NF₃ is not suited, after-treatment of the FC-containing exhaust cannot be dispensed with, but can be made significantly simpler and cheaper. Instead of high-temperature combustion, the FCs can be destroyed in a small unit by a high density plasma (HDP). If water vapour is added to the exhaust gases, FC destruction removal efficiencies of approx. 99% are achieved (SEMATECH 1999). The small unit (30x30x15 cm) is mounted directly behind the etching chamber. It costs about 75% less than an FC afterburner, and its operating costs are several times lower.³⁹ The exhaust gases exiting the downstream plasma chamber can be captured in unheated dry bed absorbers and disposed of (Kopatzki/Guerin 1999). (The HDP unit is not suited for CVD cleaning exhausts, as it cannot cope with the much larger quantities of reaction products arising compared to wafer etching.)

5.2 Assumptions for Emissions Reduction by 2010 from 1995 Baseline

Scenario II explores the lower limit of FC emissions by the year 2010. It is based on the assumption of a swift transition to NF_3 technology, comprising five specific assumptions for cleaning and five for etching.

³⁸ In July 1998, Applied Materials stated the number of chambers equipped worldwide with Remote Plasma Clean at that time to be 151 (Conners/Raoux 1998).

³⁹ According to manufacturers, the purchasing costs of a unit are 15,000 USD and its annual operation costs about 5000 USD (SEMATECH 1999).

Chamber cleaning

- 1. Total FC usage for cleaning rises annually as in Scenario I, namely by 11%. Total consumption in 2010 is equal to that in Scenario I. However, its composition in 2010 differs, namely 70% NF₃ and 30% PFCs (C₂F₆, CF₄, C₃F₈).
- 2. From the year 2000 onwards, <u>new</u> fabrication facilities use instead of C_2F_6 and other PFCs only NF₃ with upstream dissociation in microwave plasma. Utilization removal efficiency (URE) in upstream plasma dissociation figures 98%. This is so much more than in the plasma in the main process chamber (62%) that downstream exhaust treatment (burner) can be dispensed with.
- 3. In the facilities (existing facilities) installed prior to 2000, PFCs continue to be used. Existing facilities are not retrofitted to the new NF₃ technology (upstream plasma dissociation).
- 4. All facilities installed prior to 2000 retain until 2008 the downstream FC exhaust gas treatment system (burner) saturation rate of the year 1998, namely 32.5%.
- 5. From 2009 to 2010, 100% of the systems installed prior to 1996 and 75% of those installed in 1996 (existing fabrication facilities) are taken out of service and replaced by facilities with upstream NF₃ plasma dissociation.

1. Wafer etching

- The FCs used are the same as in Scenario I, namely SF₆, CHF₃ and CF₄ (partially). Their quantities develop as in Scenario I, with annual growth rates of 6% from 1998 onwards. Their total consumption in 2010 is the same as in Scenario I.
- 2. From the year 2000 onwards, <u>new</u> facilities all have downstream FC exhaust gas decomposition systems, be it in water vapour assisted HD plasma or a comparably effective system.
- 3. In the facilities (existing facilities) installed prior to 2000, the same FCs continue to be used (SF₆, CHF₃ and CF₄ partially).
- 4. All facilities installed prior to 2000 retain until 2008 the downstream FC exhaust gas treatment system (burner) saturation rate of the year 1998, namely 32.5%. Facilities without FC exhaust treatment are not retrofitted to the plasma assisted downstream FC breakdown technology obligatory for new facilities.
- 5. From 2009 to 2010, 100% of the systems installed <u>prior to 1996</u> and 75% of those installed <u>in 1996</u> (existing fabrication facilities) are taken out of service and replaced by facilities with plasma assisted downstream FC breakdown technology.

Under these conditions, the 2010 reduction target "10% less FC emissions than in 1995" is achievable.

5.3 Reduction Scenario Until 2010

In Scenario II, global warming FC emissions drop to less than 0.3 million t CO_2 equivalent by the year 2010. This is mainly attributable to the switch from 2000 onwards to the new NF₃-based chamber cleaning technology. If the ten conditions of the reduction scenario are met, FC emissions only rise until 1999 during the 15 years of the 1995-2010 period. They then remain relatively constant around 70 t until 2008, despite annual growth in FC usage by 10%. From 2009 onwards, they drop by more than half, because the highly emissive existing facilities built before 1997 are replaced

Table 83: 1995-2010 FC emissions from semiconductor manufacturing in reduction Scenario II				
	1995	2000	2005	2010
SF ₆	4	6	6	2
CHF₃	3	4	4	1
CF ₄ *	12	27	27	11
C_2F_6	11	31	28	11
NF ₃	1	1	3	5
C_3F_8	0	1	2	3
Total	2026	70	70	33
Ems. in mill. t CO ₂ equiv.	0.316	0.664	0.658	0.295

by new ones. Table 83 shows the emissions of the individual FCs for the years 1995, 2000, 2005 and 2010.

* Untransformed process gas plus new formation by plasma decomposition of C₂F₆ and C₃F₈.

Total FC consumption is the same in the reduction scenario as in the business-asusual scenario: 60 t for etching and 320 t for cleaning. Nonetheless, emissions in the reduction scenario only amount to 33 t in the year 2010 instead of 203 t (see Table 83). This is mainly due to the drop in C_2F_6 and CF_4 emissions from cleaning from 166 to 22 t, thanks to the switch to NF₃ technology. Due to the highly effective upstream plasma dissociation, NF₃ emissions only amount to 5 t, which is only 2 t more than in Scenario I.

The PFCs C_2F_6 and CF_4 continue to make up the bulk of emissions in the reduction scenario. With 11 t of each gas in the year 2010, these make up two thirds of total FC emissions (3 t of the CF₄ emissions come from etching). However, compared to the business-as-usual scenario the absolute emissions of the two gases are low.

In Scenario I, C_2F_6 is the main gas in cleaning, with 210 t consumption (2010). Of this, 101 t are emitted directly, with a further indirect emission of 10 t CF₄.

In Scenario II, 267 t NF $_3$ are consumed for chamber cleaning. Thanks to highly efficient upstream plasma dissociation, only 5 t of this are emitted.

Emissions of the etching gases SF_6 , CHF_3 and CF_4 (partially), amounting in Scenario I to 34 t in 2010, also drop in Scenario II - to 6 t. This is above all due to exhaust breakdown in downstream HD plasma.

Most FC emissions in Scenario II, ca. 20 t, come from existing facilities built between 1996 and 1999. These facilities present an opportunity for even greater emissions reductions than in the present study. The assumption that by 2010 only such facilities are decommissioned and replaced which went into operation prior to 1997 is very conservative. A lifetime of 15 years is very long for plasma facilities in the semi-conductor industry. Their manufacturers state life cycles of 8 years as being normal (Applied Materials 1999). If existing facilities are replaced by new ones earlier than assumed in the present study, total FC emissions could drop to less than 10 t by 2010.

In Scenario II, the global warming contribution in the year 2010 figures 0.295 million tonnes CO_2 equivalent. This is 6.5% less than in the baseline year 1995 (0.316 mill. t).



Diagram 39 shows the different trajectories of the two scenarios in terms of FC emissions, Diagram 40 in terms of CO₂ equivalent emissions.

Diagram 39: 1995-2010 FC emissions from semiconductor manufacturing in t FC per annum, in Scenarios I and II. If, in accordance with the assumptions of reduction Scenario II, highefficiency plasma technologies are applied from 2000 onwards, then, in contrast to businessas-usual Scenario I, emissions no longer rise, but even drop from 2009 onwards (when old facilities begin to be replaced) to the levels of 1995.



Diagram 40: 1995-2010 FC emissions from semiconductor manufacturing in million t CO_2 equivalent per annum, in Scenarios I and II. In business-as-usual Scenario I, emissions rise by 2010 to more than 1.8 million t CO_2 equivalent. Under the conditions of reduction Scenario II, in contrast, they remain constant from 2000 onwards, then to drop until 2010 to 0.295 million t - less than the baseline level of 1995.

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Characteristics of Fluorinated Compounds

GWP values of the fluorinated compounds (FCs) treated in the study (excluding chlorinated substances)				
Substance	Empirical formula	Name	GWP 100	
Perfluorocarbons				
		1/	6500	
		116	0300	
		218	<u> </u>	
Hydrofluorocarbo	$\frac{1}{10000000000000000000000000000000000$	210	7000	
Tyuronuorocarbo		23	11700	
		23	650	
		125	2800	
		13/12	1300	
		1/32	3800	
		1522	1/0	
		132a 227	2000	
		221 236fa	2900 6300	
		2301a	820	
		2451a 365mfc	810	
		43-10moo	1300	
HEC blonds		43-101166	1300	
	1/32/125/13/2	1010	3260	
	(52/1/1/4)	404A	3200	
	32/125/13/2	407C	1525 5	
	(23/25/52)	4070	1323.5	
	32/125	410A	1725	
	(50/50)			
	125/143a	507	3300	
	(50/50)			
	404A/507	404A/507	3268	
	(80/20)			
Others		1	1	
	SF ₆		23900	
	NF ₃		8000	
	C ₄ F ₉ OCH ₃	HFE 7100	500	
	÷	•	•	

Explanation

The term "reduction potential" in this study is used in the sense of a "<u>relative</u> potential". This does not characterise the technically possible, but rather the potential that can be realized in a short term first step.