

**REPORT PREPARED WITH SUPPORT FROM**

**US EPA and ADEME**

**Determination of Comparative HCFC and HFC Emission  
Profiles for the Foam and Refrigeration Sectors until 2015**

**PART 3: Total Emissions and Global Atmospheric  
Concentrations**

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## **1. Introduction**

The HCFCs and HFCs considered in this report are not just used in refrigeration, air conditioning and to blow closed-cell plastic foams, they are also used in some of the other historical applications of CFCs. Thus there are continuing uses as aerosol propellants, solvents and in open cell foam blowing. Although the quantities used in these applications are markedly less than had been the case for CFCs, for some of the compounds, notably HFC-152a, they are a significant part of the emission spectrum. The method by which background atmospheric concentrations of trace gases are calculated from the balance between their addition to the atmosphere, from emissions, and their removal, for example by chemical reaction, are described in Annex 2 to this part of the report. Clearly, in order to be able to calculate atmospheric concentrations, estimates of the total emissions are required.

## **2. Prompt Emissions**

Releases from refrigeration and air conditioning equipment and from the manufacture and use of closed cell plastic foams take place over an extended period of time. By contrast, emissions from the remaining uses are prompt; the most obvious example of this is the aerosol spray can, the functioning of which relies on release of the propellant into the atmosphere. Solvent applications also fall into this category because, even in "closed" systems, there are losses into the atmosphere from the small amount of solvent adhering to the surface of the items that are cleaned. In some foam applications the blowing agent escapes rapidly, generally when the foam cell structure is open and interconnected. For aerosols and solvents, 50% emission is estimated for the year of production of the fluorocarbon and 50% in the following year; for open cell foams the emissions are even faster, with 83% occurring in the production year and the remainder in the year following (AFEAS, 2003).

### **2.1 Global Database of Historic Emissions**

For the CFCs and HCFCs studied here, calculated prompt emissions were explicitly recorded in the AFEAS database as the total for all countries excluding India, China, Korea and Russia (AFEAS, 2003). Consumption in these countries is reported in the UN database as the aggregates separately of all CFC compounds and all HCFCs in any potentially dispersive end use. The AFEAS data for individual compounds and end uses were augmented by the emissions calculated for these additional countries in the way described in McCulloch *et al.* (2001 and 2003). This formed the historic global database for CFCs and HCFCs.

Historically HFCs have been produced only in the countries reporting into the AFEAS database, so that the values there for HFC-134a are global. Each other HFC was treated as a special case in the manner described below.

### **2.2 Predicted Emissions**

Predictions from 2002 onwards were based on extrapolation of the historic trends of the global data. The calculations were performed individually for each compound using the most appropriate scaling parameter as described below and the data used are tabulated and presented graphically in Annex 1 of this part of the report.

## **CFC-11**

This is used in aerosol propellants, open cell foam blowing and as a solvent, in addition to the major use as a closed cell foam blowing agent which has been described in the section of this report on foams. In addition there are fugitive emissions from production.

Calculated using the data in AFEAS (2003) and UNEP (2003), using the methodology in McCulloch *et al.* (2001), the sum of emissions from all prompt categories was over 110000 tonnes/year in 1990 but fell rapidly in the period up to 1995 to reach 25000 tonnes/year. Since then, the rate of fall has slowed and emissions have now levelled at approximately 15000 tonnes/year. The time series of historic emissions is shown in Figure 1 of Annex 1.

In view of the apparent stability of this emission rate, constant values were projected through to 2015 at the rate of 15400 tonnes/year in Scenario 1, 13200 tonnes/year in Scenario 2 and 8800 tonnes/year in Scenario 3. The differences reflect the assumed cessation of supply to aerosols and thereafter to both aerosols and open cell foam blowing, in scenarios 2 and 3, respectively. The values for historic and extrapolated emissions are shown in Table 1 of Annex 1.

## **CFC-12**

Like CFC-11, this is used in aerosol propellants and open cell foam blowing but its major use is as a refrigerant, which has been described in the section of this report on refrigeration and air conditioning. There are also fugitive emissions from production.

From a similar calculation to that for CFC-11, the total emissions from prompt categories was close to 90000 tonnes/year in 1990 but fell rapidly in the period to 1995 to reach just under 20000 tonnes/year. Thence it has stayed constant, mainly due to growth in unspecified "other" uses, as shown in Figure 2 of Annex 1.

Emissions were extrapolated to 2015 at constant rates: 18400 tonnes/year, in Scenario 1, 14700 tonnes/year in Scenario 2 and 13000 tonnes/year in Scenario 3, on the same basis as for CFC-11. See Table 2 of Annex 1.

## **CFC-115**

According to AFEAS (2003), there are no emissions in the prompt category; most CFC-115 is used as a refrigerant.

## **HCFC-123**

No emissions in the prompt category were assumed for this work. Due to the small number of producers, HCFC-123 cannot figure in the AFEAS database, so that there are no producers' data on end uses. However, no significant uses that might give rise to prompt emissions are known and all emissions in the calculations here were from use as a refrigerant.

## **HCFC-124**

The small quantity used in promptly emissive categories (mainly to blow open cell foam) peaked in 1997, but subsequently dropped to less than 500 tonnes/year (AFEAS, 2003). Since use of HCFCs in foams has now ceased in the EU and is regulated in other parts of the world, it is not likely that these emissions will grow and so extrapolations were made at constant rates: 500, 250 and 0 tonnes/year, for Scenarios 1 to 3, respectively. See Table 3 and Figure 3 of Annex 1.

## **HCFC-141b**

Prompt emissions are mainly from use as a solvent and in blowing open cell foam, and the values from AFEAS (2003) were augmented by UNEP (2003) records of total HCFC consumption split between the HCFCs (22, 141b and 142b) and end uses, as described in McCulloch *et al.* (2003) to provide a global estimate. AFEAS (2003) data were used exclusively for the medium term emissions (from refrigeration).

Only one scenario was extrapolated, as a linear decline using the separate trends for prompt and medium term emissions in the period 1995 to 2001. Table 4 and Figure 4 of Annex 1 show the values and trends.

## **HCFC-142b**

Prompt emissions arise from use in blowing open cell foam and there are some medium term emissions from refrigeration. The rates were calculated in the same way as for HCFC-141b, described above.

Although prompt emissions went through a maximum in 1992, in recent years the values have been erratic, varying around a mean of about 860 tonnes/year. For the single scenario considered, prompt emissions were held constant at the average rate from 1997 onwards. For the medium term emissions there is a far clearer trend, which was extrapolated linearly to 2015. Table 5 and Figure 5 of Annex 1 show the values used.

## **HCFC-22**

Historically, there was almost compound growth in prompt releases up to 1989. Thenceforward, emission rates have been highly erratic. The prompt emissions from use mainly in open cell foam were adjusted to reflect global use of HCFC-22 in the same way as for HCFCs 141b and 142b. Table 6 and Figure 6 of Appendix 1 shows the values and also the large influence of fugitive emissions from production. In addition to the dispersive uses described in this report, HCFC-22 is a feedstock for fluoropolymers (which is not controlled) and fugitive emissions are expected to increase as the total quantity produced increases. In view of the recent erratic history, dispersive uses were extrapolated at three constant levels for the three scenarios. Scenario 1 used the mean of the maximum rates (30000 tonnes/year); for Scenario 2, a constant median rate of 20000 tonnes/year was adopted and, for Scenario 3, the level was set at the value where long term trend had finished (10000 tonnes/year). Fugitive emissions were added in at 2.5% of total production (Midgley and Fisher, 1993).

## **HFC-125**

No prompt emissions are recorded in the UN database (UNFCCC, 2004) and an arbitrary allowance of 100, 75 and 50 tonnes/year was made for Scenarios 1 to 3, respectively.

## **HFC-134a**

Prompt emissions arise from use as an aerosol propellant (both for medical and technical aerosols), from open cell foam blowing and from use in one component foam packs (gap-fillers). Figure 7 of Annex 1 shows growth since 1990 but recently there has been an apparent fall in the trend. For Scenario 1, the long term trend was extrapolated linearly to give a maximum rate. In the case of Scenario 2, the more recent trend was extrapolated linearly and was assumed to represent the median so that, in Scenario 3, the trend is less than Scenario 2 by the same difference as that between Scenarios 1 and 2. The values obtained are given in Table 7 of Annex 1.

## **HFC-143a**

No prompt emissions are recorded in the UN database (UNFCCC, 2004) and an arbitrary allowance of 70, 50 and 30 tonnes/year was made for Scenarios 1 to 3, respectively.

## **HFC-152a**

Prompt emissions from use in aerosol propulsion (mainly technical sprays), open cell foam blowing and one component foam packs far outweigh the other uses. Since there are few significant producers of HFC-152a, there are no data on production and consumption but the emissions reported to UNFCCC (2004) by EU member states match the confidential database maintained for regulators by EU industry (personal communication, Cefic, 2004). These data show that over 80% of emissions of HFC-152a in the EU come from prompt categories and that their growth is linear.

There are significant emissions from replacement of VOCs in aerosols in the U.S.A. but these are not reported explicitly to UNFCCC (2004). These considerations have meant that HFC-152a emissions have been treated differently from the other fluorocarbons described here. HFC-152a emissions have been fitted to the atmospheric measurements. Montzka, Fraser *et al.* (2003) report atmospheric concentrations of  $1.3 \text{ pmol mol}^{-1}$  in 1998 and  $1.7 \text{ pmol mol}^{-1}$  in 2000. In order to maintain these absolute values and the growth rate, prompt emissions growing at a rate of 1250 tonnes/year would be required. Starting in 1990, values from this trend have been used for the prompt emissions in order to be able to place emissions from other sources into context.

## **HFC-227ea**

Prompt emissions arise from use in medical aerosols and the confidential industrial data (personal communication, Cefic, 2004) show long term relatively slow growth. The trend line for this data is given in Table 8 and Figure 8 of Annex 1. Medium term emissions come from use in fire extinguishing systems and show more rapid growth over a shorter time span. Again the trend line only is shown.

**HFC-245fa**

There is no history of emissions and no allowance has been made for future prompt emissions.

**HFC-32**

All use of this material is in refrigeration (UNFCCC, 2004).

**HFC-365mfc**

All use of this material is in closed cell foams (personal communication, Cefic, 2004).



### 3. Global Emissions from all Sources

#### 3.1 Historical Data

The total emissions comprise the releases from refrigeration, air conditioning, foam blowing and all of the promptly emissive categories estimated in this report. Historical data are shown in Table 3.1 for the ozone depleting substances (CFCs and HCFCs) and in Table 3.2 for the HFCs.

**Table 3.1** Historic Emissions of Ozone Depleting Substances (tonnes/yr)

Year	CFC-11	CFC-12	CFC-115	HCFC-123	HCFC124	HCFC-141b	HCFC-142b	HCFC-22
1990	145275	222010	9578	0	0	12	2098	148817
1991	106094	190132	9838	65	27	691	3453	151897
1992	80928	177249	10136	152	124	3897	6228	159170
1993	66697	166420	10303	456	237	10916	5514	158807
1994	58832	153609	10315	1225	389	18358	6377	168850
1995	52081	146446	9774	2099	1552	25430	9458	179703
1996	54936	137607	9588	2750	3310	24675	10276	194682
1997	48674	135664	10303	3404	4093	23721	10265	196222
1998	42558	132317	10581	3656	2655	26664	9926	201117
1999	41102	125503	10269	3869	2775	26560	10368	216737
2000	42060	122044	9938	4155	2918	25583	11057	236220
2001	43017	119007	10005	3922	3281	28073	11808	247892

**Table 3.2** Historic Emissions of HFCs (tonnes/yr)

Year	HFC-125	HFC-134a	HFC-143a	HFC-152a	HFC-227ea	HFC-245fa	HFC-32	HFC-365mfc
1990	0	175	0	12	0	0	0	0
1991	0	239	0	1265	0	0	0	0
1992	5	589	35	2541	0	0	0	0
1993	26	3425	169	3866	185	0	0	0
1994	76	8169	498	5190	182	0	0	0
1995	200	17483	1114	7345	100	0	0	0
1996	472	30015	1966	8853	437	0	0	0
1997	971	37442	3045	10257	408	0	0	0
1998	1900	43052	4619	11664	928	0	5	0
1999	3240	57607	6652	13223	1354	0	45	0
2000	5153	73681	9184	15198	1951	0	229	0
2001	7491	79863	12031	15921	1645	3	392	0

It is interesting to compare these estimates of the emissions with those in the literature for CFCs 11 and 12, HCFC-22 and HFC-134a (Table 3.3). These values were developed directly from the sales and consumption activity (for all categories) reported in AFEAS (2003), UNEP (2003) and are catalogued in the Global Emissions Inventory Activity, available at <http://weather.engin.umich.edu/geia/>. They are used in Annex 2 of this report (the literature survey) to demonstrate the methodology for verification of emissions inventories using atmospheric measurements.

There are some clear differences, notably that the literature values are generally higher in the early 1990s and begin to approach the values in this report only towards the end of the decade. The difference quoted in Table 3.3 is the average root mean square difference between the data sets calculated as a percentage of the mean of the data set. The effect of such differences on verification by atmospheric measurements will be examined more fully in the section of this report on atmospheric concentrations.

**Table 3.3** Historic Emissions Estimates from literature values (tonnes/yr)

Year	CFC-11	CFC-12	HCFC-22	HFC-134a
1990	257663	367100	216553	61
1991	229984	336600	229030	245
1992	212071	319900	237219	834
1993	198974	300500	239036	3605
1994	118528	277000	246326	9057
1995	106178	255500	251513	19779
1996	100609	233100	266048	32000
1997	92385	207700	259441	41861
1998	84719	182000	269403	53866
1999	79376	155200	279514	69826
2000	74768	134100	285842	85260
2001			293230	97547
Difference	19.9%	34.8%	8.4%	7.2%
Source	(a)	(b)	(b)	(b)

(a) McCulloch *et al.* (2001)

(b) McCulloch *et al.* (2003)

### 3.2 Forecasts for Future Emissions

The combined scenarios for future emissions were calculated in the same way as the historic data by summation of the contributions from individual end uses described in the first sections of this report. Scenario 1 is intended to illustrate the outcome from continuing with current practices, with no improvement to recovery efficiency but taking into account the effects of current regulations. For Scenario 2, improvements are introduced to reduce equivalent CO<sub>2</sub> emissions of fluorocarbons: using more reliable components to improve system leak tightness; obtaining better recovery efficiency at servicing and end of life and introducing recovery to sectors where it was not done before; promoting technologies to reduce the refrigerant charge or blowing agent use and switching to lower GWP materials. Scenario 3 envisages a somewhat more draconian approach, with every opportunity taken to implement the measures in Scenario 2.

In some cases, only one scenario has been developed. For HCFCs 141b and 142b, there was no basis to develop other scenarios because the major components of future values were founded on simple extrapolations. It could be argued that, in Scenarios 2 and 3, the deployment of these HCFCs should be drastically reduced. However, such reductions are regulated only in the developed world and the developing countries are not constrained until 2015 and have every incentive to maximise their use of HCFCs by then.

HFCs 227ea, 245fa and 365mfc present a somewhat different case. For these compounds there is so little historical data that the provision of three scenarios strained credibility. A single scenario representing the environmentally "worst case" for maximum emissions was therefore presented.

Tables 3.4, 3.5 and 3.6 show Scenarios 1, 2 and 3 for the CFCs and HCFCs and, similarly, Tables 3.7, 3.8 and 3.9 show the scenarios for HFCs. The data in these tables has been used in subsequent sections of this report to calculate atmospheric concentrations.

**Table 3.4** Forecast Emissions of ODS under Scenario 1 (tonnes/yr)

Year	CFC-11	CFC-12	CFC-115	HCFC-123	HCFC124	HCFC-141b	HCFC-142b	HCFC-22
2002	42638	116640	10475	4151	3368	25816	10521	271626
2003	42238	116240	10871	4340	3205	22129	10761	286242
2004	41710	115854	12092	4385	2848	20525	11237	310042
2005	40961	109080	11582	4013	2611	19718	10917	332319
2006	40202	103164	9873	3497	2515	19108	10877	351859
2007	39220	99819	9476	3064	2461	18967	11335	377876
2008	38529	96742	8785	2649	2435	18891	11812	404045
2009	37658	88867	7857	2454	2321	19341	12303	426615
2010	36863	79620	6630	2354	2176	19797	12039	449811
2011	36114	71190	5545	2215	1997	20471	7836	458446
2012	35227	62940	4568	2357	1882	21034	7814	472388
2013	34697	53972	3537	2412	1776	20812	7789	485910
2014	34111	46187	2736	3292	1679	20795	7768	491156
2015	33552	40321	2060	5634	1558	20817	7746	492929

**Table 3.5** Forecast Emissions of ODS under Scenario 2 (tonnes/yr)

Year	CFC-11	CFC-12	CFC-115	HCFC-123	HCFC124	HCFC-141b	HCFC-142b	HCFC-22
2002	41791	108631	10196	3987	3135	-	-	253922
2003	40697	105718	10435	4063	2851	-	-	263721
2004	39485	102902	11492	3985	2396	-	-	281832
2005	38062	95171	10888	3516	2127	-	-	297347
2006	37208	89037	9157	2945	1988	-	-	309781
2007	36141	84948	8665	2466	1890	-	-	328167
2008	35379	81344	7955	2002	1817	-	-	344688
2009	34447	73872	7058	1756	1679	-	-	358379
2010	33608	65524	5932	1598	1523	-	-	371610
2011	32818	57641	4928	1414	1351	-	-	372998
2012	31900	50018	4051	1428	1233	-	-	377595
2013	31350	41942	3122	1407	1127	-	-	381396
2014	30753	34952	2413	1865	1030	-	-	379067
2015	30195	29754	1820	3132	924	-	-	373575

**Table 3.6** Forecast Emissions of ODS under Scenario 3 (tonnes/yr)

Year	CFC-11	CFC-12	CFC-115	HCFC-123	HCFC124	HCFC-141b	HCFC-142b	HCFC-22
2002	40503	104244	9980	3873	2934	-	-	237888
2003	38233	99688	10107	3883	2544	-	-	244263
2004	35851	96201	10993	3743	2001	-	-	258233
2005	33264	87239	10286	3241	1712	-	-	269071
2006	32339	80459	8552	2660	1540	-	-	276765
2007	31212	75619	7953	2178	1412	-	-	289411
2008	30397	71245	7174	1724	1306	-	-	299005
2009	29419	63736	6247	1456	1155	-	-	305271
2010	28550	55773	5157	1261	997	-	-	310170
2011	27729	48244	4182	1046	839	-	-	305286
2012	26786	41229	3348	985	722	-	-	302519
2013	26218	34456	2509	908	619	-	-	298329
2014	25612	28610	1893	1097	524	-	-	289447
2015	25050	24335	1399	1631	425	-	-	277512

Scenarios 2 and 3 were not calculated for HCFCs 141b and 142b.

**Table 3.7** Forecast Emissions of HFCs under Scenario 1 (tonnes/yr)

Year	HFC-125	HFC-134a	HFC-143a	HFC-152a	HFC-227ea	HFC-245fa	HFC-32	HFC-365mfc
2002	9972	92403	14835	18556	2508	150	568	65
2003	12608	104369	17425	20605	3341	1133	822	358
2004	15693	120196	20438	21863	4269	1703	1146	794
2005	19003	138174	23404	23007	4890	2181	1701	923
2006	22039	152801	26218	24164	5545	2559	2227	1087
2007	24410	166021	28147	25443	6160	2813	2800	1210
2008	26891	179838	30166	26694	6775	3071	3424	1334
2009	29849	197049	32532	27856	7386	3336	4127	1453
2010	33151	216328	35283	28953	7992	3607	4878	1566
2011	36602	233897	38188	30002	8587	3879	5569	1670
2012	39493	250065	41009	31246	9166	4151	6323	1757
2013	43461	266695	43693	32415	9721	4420	7285	1820
2014	47568	284379	46286	33482	10241	4683	8964	1849
2015	50944	300668	48937	34399	10713	4933	9842	1829

**Table 3.8** Forecast Emissions of HFCs under Scenario 2 (tonnes/yr)

Year	HFC-125	HFC-134a	HFC-143a	HFC-152a	HFC-227ea	HFC-245fa	HFC-32	HFC-365mfc
2002	9573	86652	14243	18510	-	-	547	-
2003	11896	94460	16386	20538	-	-	811	-
2004	14556	102102	18805	21785	-	-	1162	-
2005	17334	115725	21062	22918	-	-	1745	-
2006	19693	126088	23045	24059	-	-	2250	-
2007	21350	135101	24153	25321	-	-	2771	-
2008	23054	144375	25266	26554	-	-	3355	-
2009	24871	155561	26370	27706	-	-	3995	-
2010	26790	167563	27371	28799	-	-	4896	-
2011	28674	176367	28446	29851	-	-	5649	-
2012	30374	185133	29340	31093	-	-	6422	-
2013	31965	192990	30038	32262	-	-	7271	-
2014	33843	201714	30580	33330	-	-	8551	-
2015	35023	208498	31017	34252	-	-	9222	-

Scenario 2 was not calculated for HFCs 227ea, 245fa or 365mfc

**Table 3.9** Forecast Emissions of HFCs under Scenario 3 (tonnes/yr)

Year	HFC-125	HFC-134a	HFC-143a	HFC-152a	HFC-227ea	HFC-245fa	HFC-32	HFC-365mfc
2002	9260	77567	13789	18476	-	-	530	-
2003	11309	84601	15586	20489	-	-	771	-
2004	13567	94020	17534	21728	-	-	1083	-
2005	15822	104420	19226	22854	-	-	1595	-
2006	17558	111314	20542	23983	-	-	2008	-
2007	18589	116407	21019	25233	-	-	2413	-
2008	19578	120346	21441	26454	-	-	2848	-
2009	20513	126571	21597	27602	-	-	3413	-
2010	21520	132334	21332	28693	-	-	4471	-
2011	22365	135919	21153	29750	-	-	5273	-
2012	22890	136283	20770	30994	-	-	5974	-
2013	23238	137401	20209	32164	-	-	6671	-
2014	23604	139287	19516	33232	-	-	7507	-
2015	23471	135766	18731	34157	-	-	7926	-

Scenario 3 was not calculated for HFCs 227ea, 245fa or 365mfc.

## 4. Atmospheric Concentrations

### 4.1 Historic Measurements

Mass balance of a long-lived trace gas in the atmosphere involves comparison of the measured concentrations in the well-mixed, background atmospheric air with values derived from the quantity of the trace gas that has been released, adjusted for the quantities that are lost from the atmosphere, for example by chemical reaction. Accurate measurements of these background concentrations are therefore a prerequisite.

Within the Atmospheric Lifetime Experiment and its successors, the Global Atmospheric Gases and the Advanced Global Atmospheric Gases Experiments (referred to collectively as AGAGE), there are time series of up to 25 years of *in situ* measurements for fluorocarbon trace gases. These are published at [ftp://cdiac.esd.ornl.gov/pub/ale\\_gage\\_Agage](ftp://cdiac.esd.ornl.gov/pub/ale_gage_Agage) and cover five sites worldwide. A somewhat shorter data record is available from the work of the Climate Diagnostics Laboratory of the U.S. National Oceanic and Atmospheric Administration (CMDL). This covers both flask samples and *in situ* measurements for some 10 sites worldwide and results are published at <http://www.cmdl.noaa.gov>. Other laboratories are active in this field but have less comprehensive geographical coverage.

A summary of the intercomparison of results from all laboratories, that was carried out in 2002 as part of the Scientific Assessment of Ozone Depletion (Montzka, Fraser *et al.*, 2003), is shown in Table 4.1. It is clear that, even with the care taken within each laboratory to lessen the uncertainty of the analyses, there are significant differences between the global average concentrations calculated from measurements. These arise not only from analytical variability and systematic differences but also from the way that the databases from different geographical locations are used to calculate a "global average" and so it is important to use a self consistent database. In this report, the data from the AGAGE record at Mace Head, Ireland, was taken to be typical of the Northern hemispherical concentrations and, for the Southern hemisphere, the Cape Grim records from the same experiment were used. The time series of measurements from other laboratories are similar but the AGAGE values are the longest from the same point using the exactly the same technique and, furthermore, are consistent with the values obtained using previous techniques at the same location, hence they have a provenance that, in some cases, spans 25 years.

Because the AGAGE data are measured using a common technique continuously from the same geographical location, background air samples can be separated from those from polluted air streams using statistical techniques (as described in Section 2.3 of Annex 2).

Concentrations were calculated from the emissions listed in Tables 3.1 and 3.2 using the two box model of the atmosphere described in Section 3 of Annex 2 and the atmospheric lifetimes and hemispherical distributions of emissions shown in Table 4.2.

**Table 4.1** Recent Globally Averaged Atmospheric Concentrations of Ozone Depleting Substances (Montzka, Fraser *et al.*, 2003)

Substance	Estimate	Concentration pmole mole <sup>-1</sup>			Growth rate pmole mole <sup>-1</sup> yr <sup>-1</sup>	Number of Laboratories
		1996	1998	2000	1999-2000	
CFC-12	High	532.4	538.4	542.9	2.3	4
	Low	523.2	529.3	534.0	1.8	
CFC-11	High	270.5	267.2	263.2	-2.0	4
	Low	265.6	263.0	260.5	-1.0	
HCFC-22	High	122.4	132.7	143.2	5.4	2
	Low	121.5	131.4	141.9	5.1	
HCFC-141b	High	5.4	9.5	13.0	1.8	3
	Low	4.3	9.1	12.7	1.7	
HCFC-142b	High	9.2	10.4	12.5	1.1	3
	Low	7.7	9.6	11.7	1.0	
HCFC-124		0.03	0.89	1.34	0.35	2

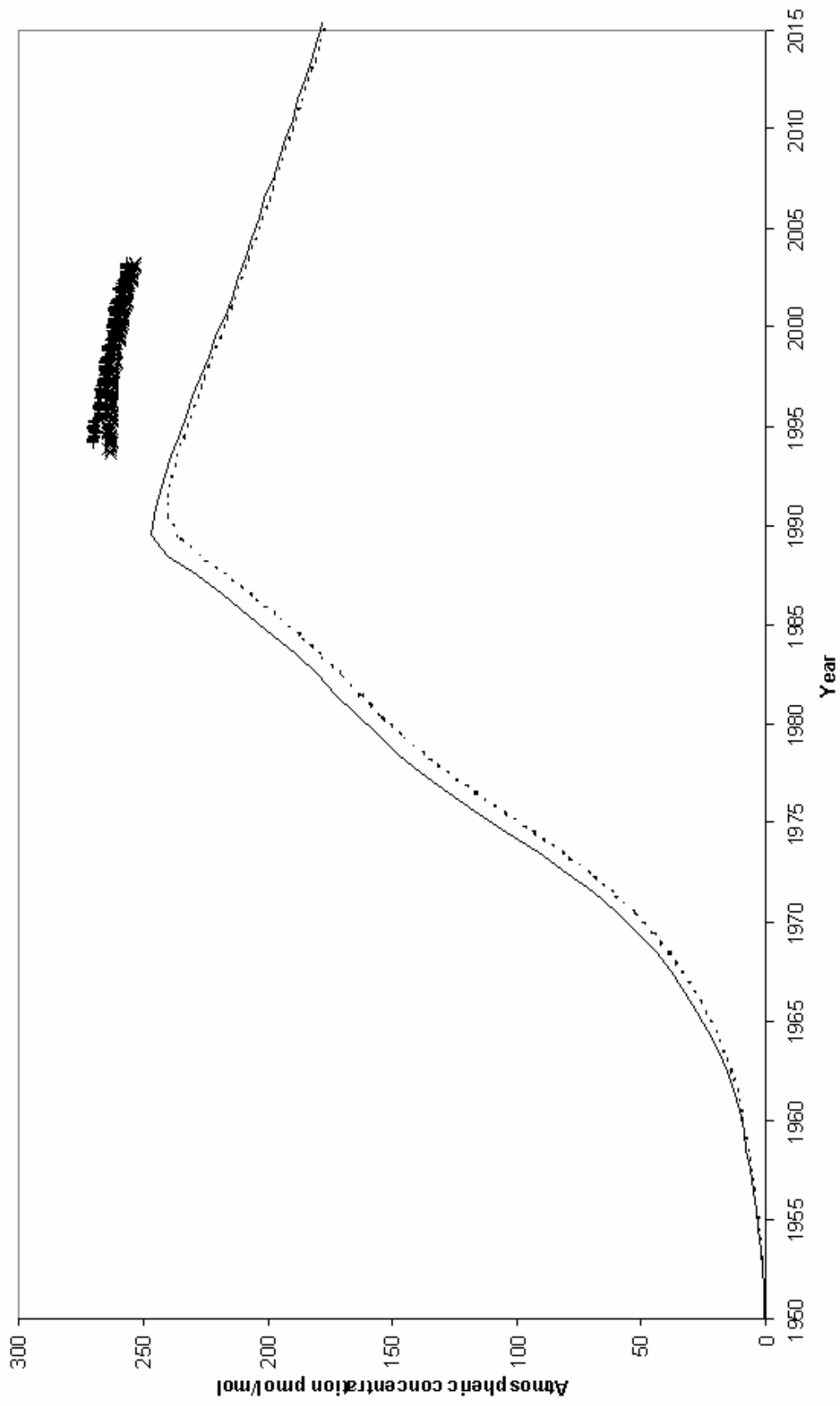
**Table 4.2.** Parameters Required for Two Box Models of Fluorocarbon Concentrations from Emissions

Substance	Atmospheric Lifetime (years) <sup>1</sup>	Distribution of Emissions (%) <sup>2</sup>					
		Scenario 1		Scenario 2		Scenario 3	
		NH	SH	NH	SH	NH	SH
CFC-11	45	89.3	10.7	89.2	10.8	89.1	10.9
CFC-12	100	89.3	10.7	89.2	10.8	89.1	10.9
CFC-115	1700	89.3	10.7	89.2	10.8	89.1	10.9
HCFC-123	1.3	95.8	4.2	95.7	4.3	95.8	4.2
HCFC-124	5.8	95.8	4.2	95.7	4.3	95.8	4.2
HCFC-141b	9.3	95.8	4.2	95.7	4.3	95.8	4.2
HCFC-142b	17.9	95.8	4.2	95.7	4.3	95.8	4.2
HCFC-22	12	95.8	4.2	95.7	4.3	95.8	4.2
HFC-125	29	96.5	3.5	96.2	3.8	95.9	4.1
HFC-134a	14	96.5	3.5	96.2	3.8	95.9	4.1
HFC-143a	52	96.5	3.5	96.2	3.8	95.9	4.1
HFC-152a	1.4	96.5	3.5	96.2	3.8	95.9	4.1
HFC-227ea	34.2	96.5	3.5	96.2	3.8	95.9	4.1
HFC-245fa	7.6	96.5	3.5	96.2	3.8	95.9	4.1
HFC-32	4.9	96.5	3.5	96.2	3.8	95.9	4.1
HFC-365mfc	8.6	96.5	3.5	96.2	3.8	95.9	4.1

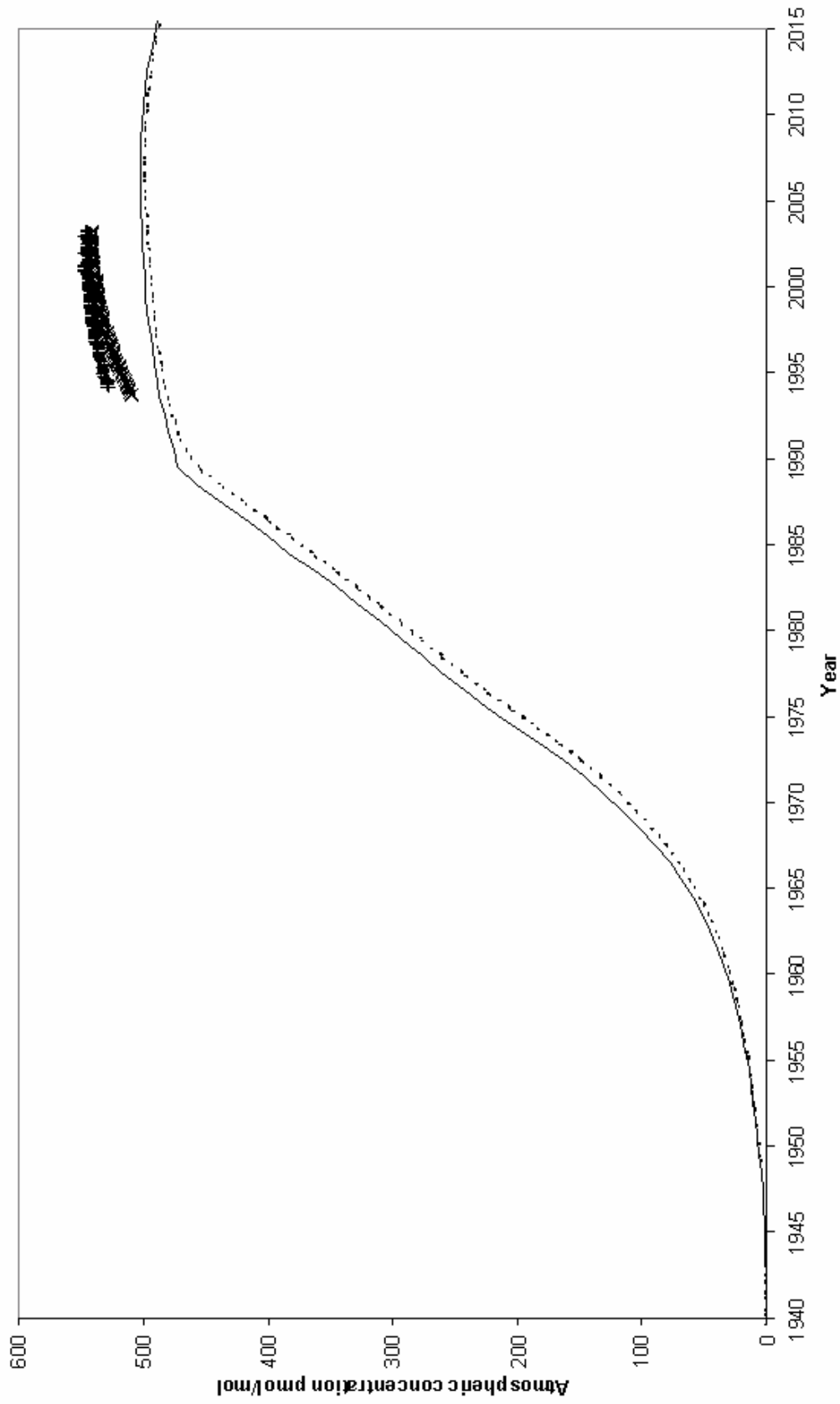
<sup>1</sup> Source - Montzka, Fraser *et al.* (2003)

<sup>2</sup> Source - This report





**Figure 1.** Atmospheric Concentrations of CFC-11. Measured data from AGAGE (2004) (Northern hemisphere as +; Southern as X), concentrations calculated as described in the text. Scenario 1 only shown, other Scenarios similar.



**Figure 2.** Atmospheric Concentrations of CFC-12. Measured data from AGAGE (2004) (Northern hemisphere as +; Southern as X), concentrations calculated as described in the text. Scenario 1 only shown, other Scenarios similar.

## **CFC-11**

The measured concentrations of CFC-11 are shown in Figure 1 as the dense series of plus signs (Mace Head measurements) and crosses (Gape Grim). Each symbol is a monthly mean of, on average, 700 separate baseline determinations and the average monthly standard deviation is 0.14%. Also shown on Figure 1 are the hemispherical concentrations calculated from emissions using the model from Section 3 of Annex 2 and the parameters in Table 4.2 above. Up to 1989 the emissions estimated in McCulloch *et al.* (2001) were used, between 1990 and 2001 the values in Table 3.1 and, subsequently, the values in Table 3.4 were used. It is clear that the data sets are not consistent with each other and that the data from this report underestimate atmospheric concentrations. Potential reasons for this are investigated later in this report.

Figure 1 shows only Scenario 1. Concentrations resulting from the other scenarios described in Tables 3.5 and 3.6 were very similar and, in view of the mismatch between historical emissions and the variation in future concentrations is not pursued further here.

## **CFC-12**

Figure 2 was constructed similarly to Figure 1, with plus signs representing monthly mean concentrations from Mace Head and crosses the Cape Grim determinations. The measurement statistics are similar to CFC-11 (700 determinations and 0.12% average standard deviation). Again, the hemispherical concentrations show a point of inflection at 1990 which coincides with the change of emissions database; in this case from McCulloch *et al.* (2003) to that shown in Tables 3.1 and 3.4.

Figure 2 shows only Scenario 1. As for CFC-11, concentrations resulting from the other scenarios described in Tables 3.5 and 3.6 were very similar and the variation in future concentrations is not pursued further here.

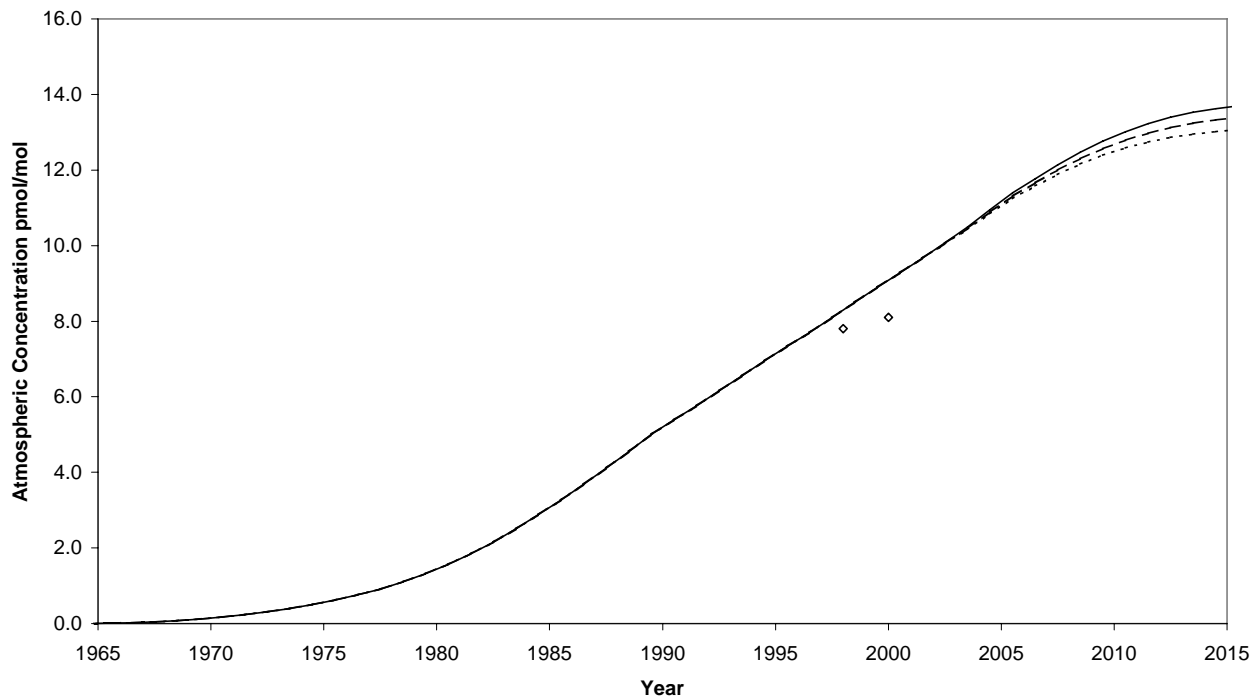
## **CFC-115**

CFC-115 does not figure in the AGAGE database and the average annual concentrations reported in Montzka, Fraser *et al.*, (2003), 7.8 pmol mol<sup>-1</sup> in 1998 and 8.1 in 2000, are shown in Figure 3. The AFEAS database carries the complete record of global production of CFC-115 up to 1989 (Fisher and Midgley, 1994) and was used to calculate concentrations here, with data from Tables 3.1 and 3.4 to 3.6 being used subsequently. There is a much less pronounced point of inflection at 1990 but the calculated concentrations exceed measurements (to be discussed later in this report).

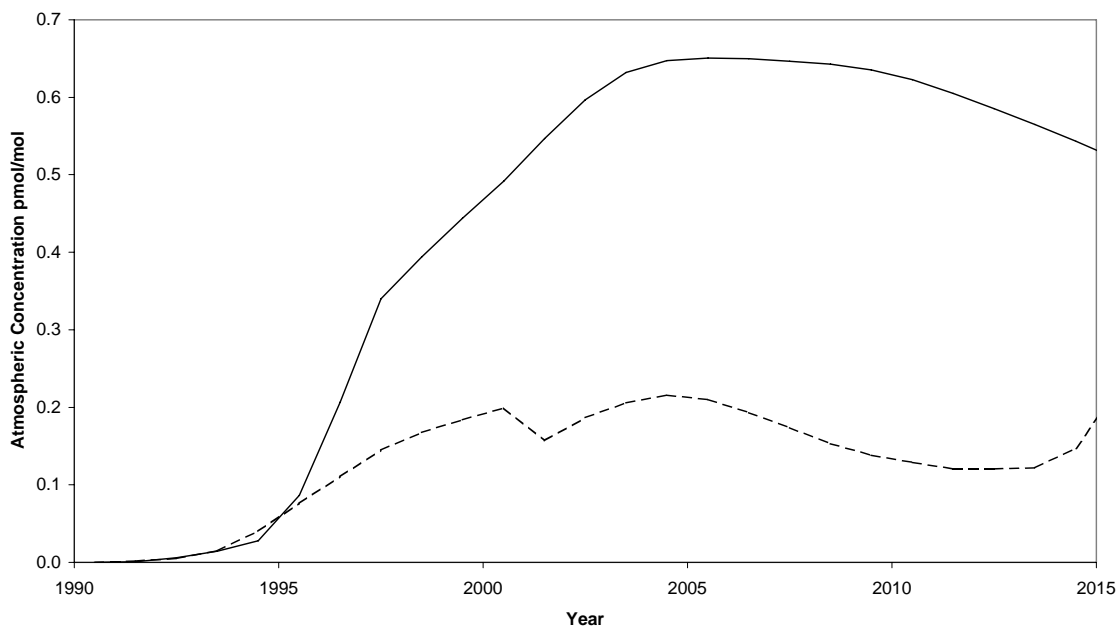
In the case of CFC-115, all scenarios are shown on Figure 3.

## **HCFC-123**

The atmospheric concentrations of HCFC-123 have not been published in the same rigorous way as the other compounds discussed here. They are thought to be very low (less than 0.5 pmol mol<sup>-1</sup>) and consistent with the concentrations shown in Figure 4 that were calculated from the data in Table 3.1.



**Figure 3.** Atmospheric Concentrations of CFC-115. Measured data (open diamonds, as global averages) from Montzka, Fraser *et al.* (2003), global average concentrations calculated as described in the text: solid line - Scenario 1, dashed line - scenario 2 and dotted line - Scenario 3.



**Figure 4.** Calculated Global Average Atmospheric Concentrations of HCFCs 123 (dashed) and 124 (solid). Scenario 1 only shown.

## **HCFC-124**

Global data from Montzka, Fraser *et al.* (2003), reproduced in Table 4.1 above shows a rapidly rising concentration in the region of  $1.3 \text{ pmol mol}^{-1}$ , somewhat higher than the concentration calculated from emissions shown in Figure 4. However, at this low level, determination of the concentration has high uncertainty and, even if production and releases of HCFC-124 were to continue (which is considered unlikely), its atmospheric concentration would not become significant compared to that of HCFC-22, for example. Consequently, no further consideration has been given to either HCFC-123 or HCFC-124.

## **HCFC-141b**

The measured concentrations of HCFC-141b are shown in Figure 5 as the series of plus signs (Mace Head measurements) and crosses (Gape Grim). Each symbol is a monthly mean of, on average, 74 separate baseline determinations and the average monthly standard deviation is 1.8%. The comparatively large difference between hemispherical concentrations is due to their rapid increase with almost all of the emissions in the Northern hemisphere. The mixing between hemispheres shows up as a delay of approximately two years between the concentration in the south matching that in the north and this is exaggerated by the steep rise in concentrations.

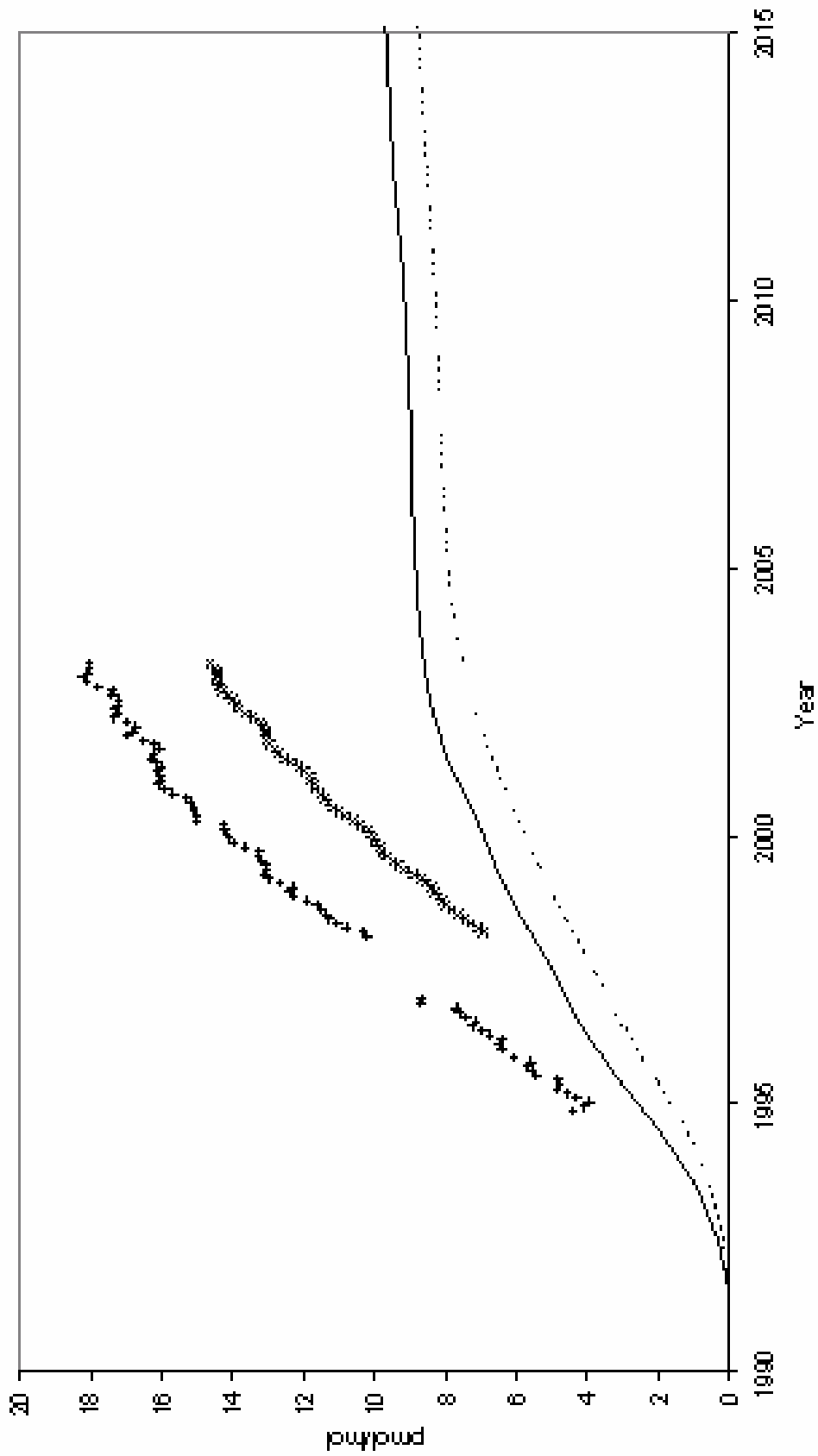
Also shown on Figure 5 are the hemispherical concentrations calculated from emissions using the model from Section 3 of Annex 2, the parameters in Table 4.2 above and the emissions of Tables 3.1 and 3.4. It is clear that the actual atmospheric concentrations are underestimated in the calculations by a large margin. Potential reasons for this are investigated later in this report.

Emissions only for Scenario 1 have been calculated.

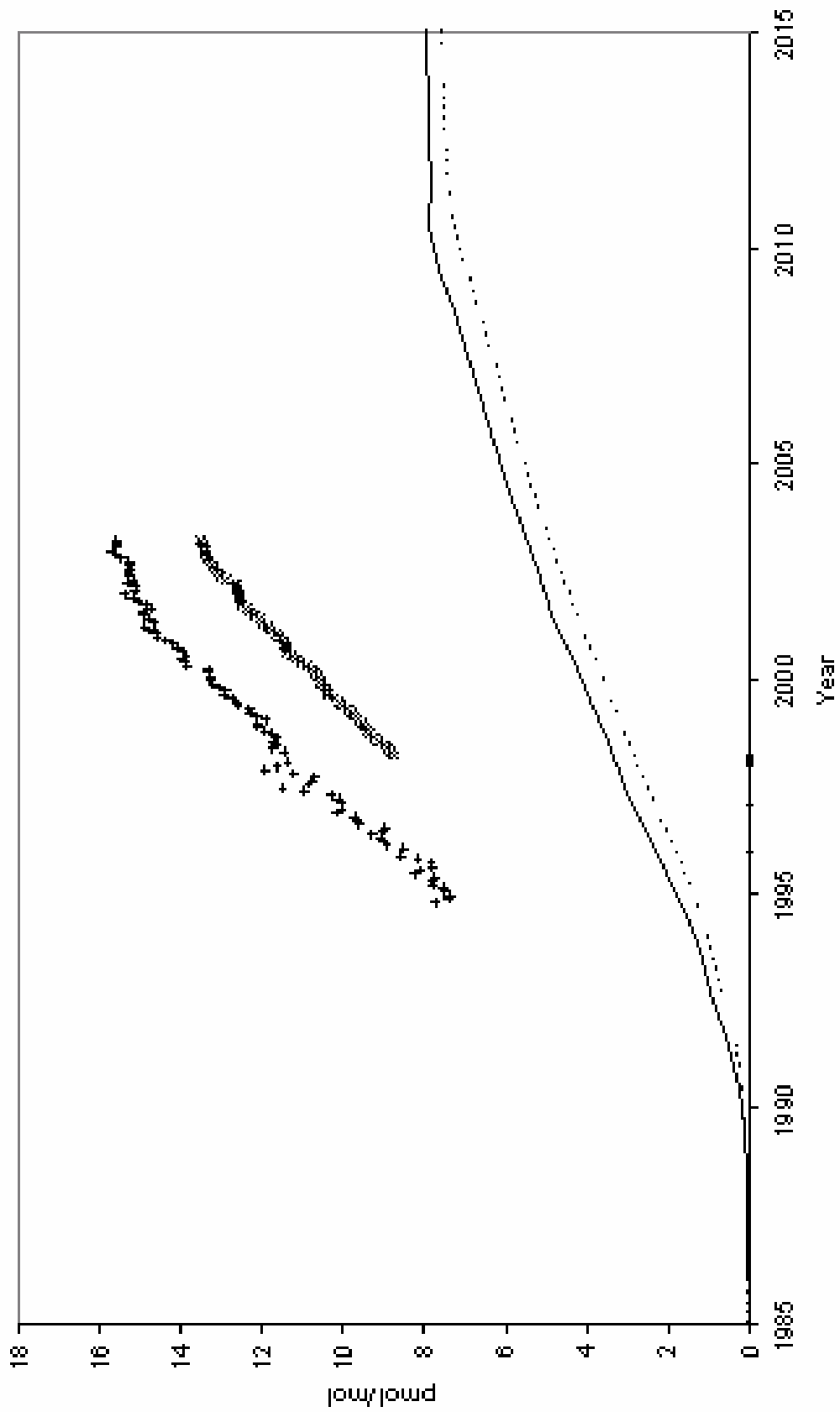
## **HCFC-142b**

Figure 6 was constructed similarly to Figure 5, with plus signs representing monthly mean concentrations from Mace Head and crosses the Cape Grim determinations. The measurement statistics are similar to HCFC-141b (84 determinations and 2.1% average standard deviation). The differential between hemispheres is similar to that for HCFC-141b, for the same reasons.

Up to 1989 the small historical emissions estimated in AFEAS (2003) were used. Subsequently, the values in Tables 3.1 and 3.4 developed in this report were used. Like HCFC-141b, there is a very large difference between calculated and measured values that will be discussed later in this work.



**Figure 5.** Atmospheric Concentrations of HCFC-141b - plus signs Mace Head data, crosses Cape Grim data - with calculated values for Northern hemisphere (solid) and Southern hemisphere (dotted). Only one scenario shown.



**Figure 6.** Atmospheric Concentrations of HCFC-142b - legend as for Figure 5, 141b, on previous page.

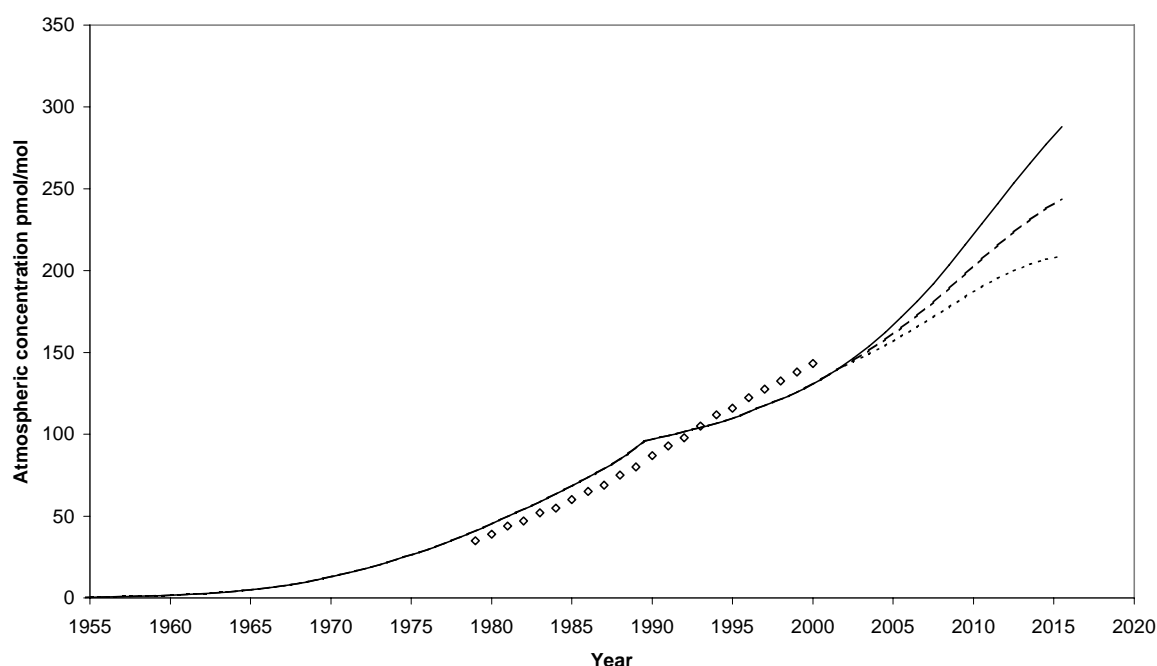
## HCFC-22

The globally averaged measured concentrations shown in Figure 7 were taken principally from Prinn *et al.* (2000), with a one year extrapolation based on the data in AFEAS (2003) and UNEP (2003). The calculated concentrations were based on McCulloch *et al.* (2003) up to 1990 and thenceforward on the emissions from this work shown in Tables 3.1 and 3.4 to 3.6.

Although there is a point of inflection in 1990 where the underlying databases change, the measured concentrations are relatively well matched by those calculated from the historic emissions data.

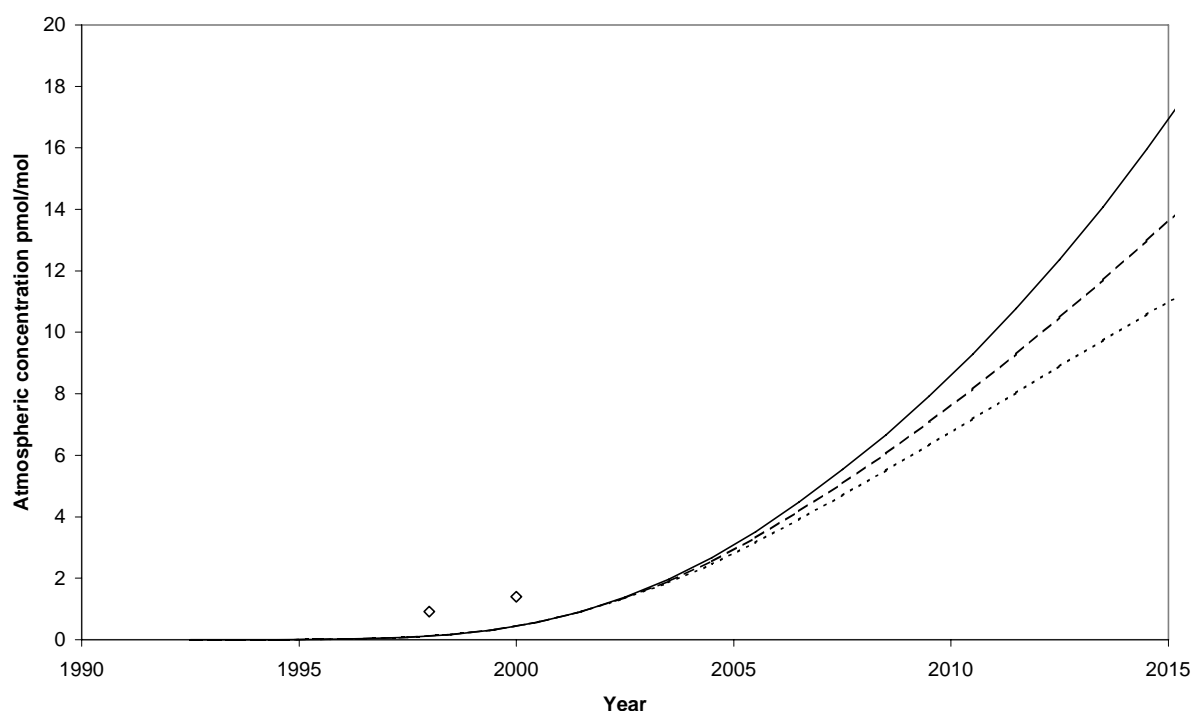
## HFC-125

Atmospheric measurements of the concentration of HFC-125 (Montzka, Fraser *et al.*, 2003) are substantially higher than those calculated from the emissions shown in Table 3.2, see Figure 8. However, the absolute magnitude of the concentrations is very low (less than 2  $\text{pmol mol}^{-1}$ ) and so the measurement errors may be substantial. There is no indication of a significant source of emissions other than refrigeration.



**Figure 7.** Atmospheric concentrations of HCFC-22. Open diamonds show the globally averaged measurements and the lines show globally averaged concentrations calculated from emission data for all three scenarios.

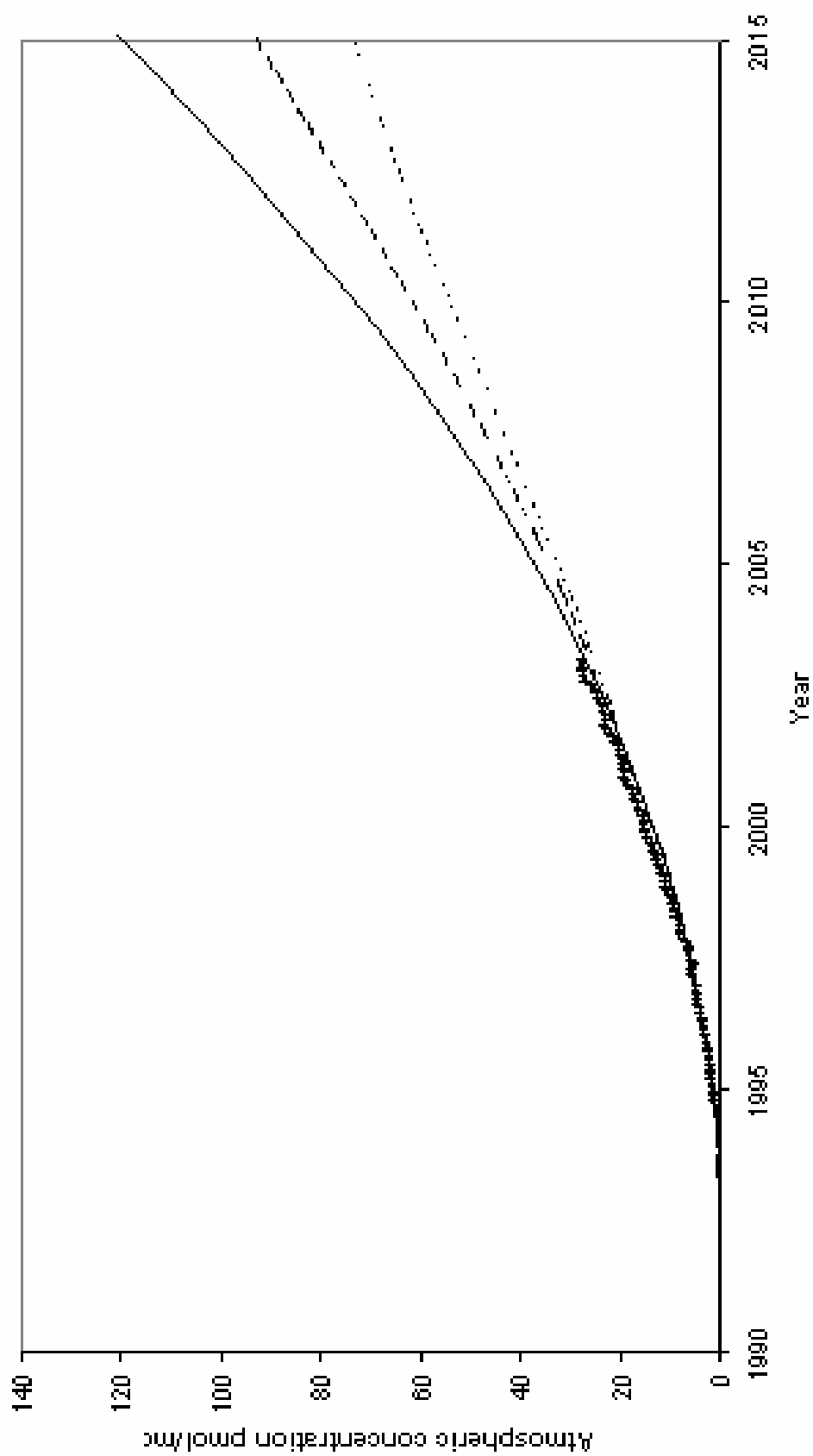




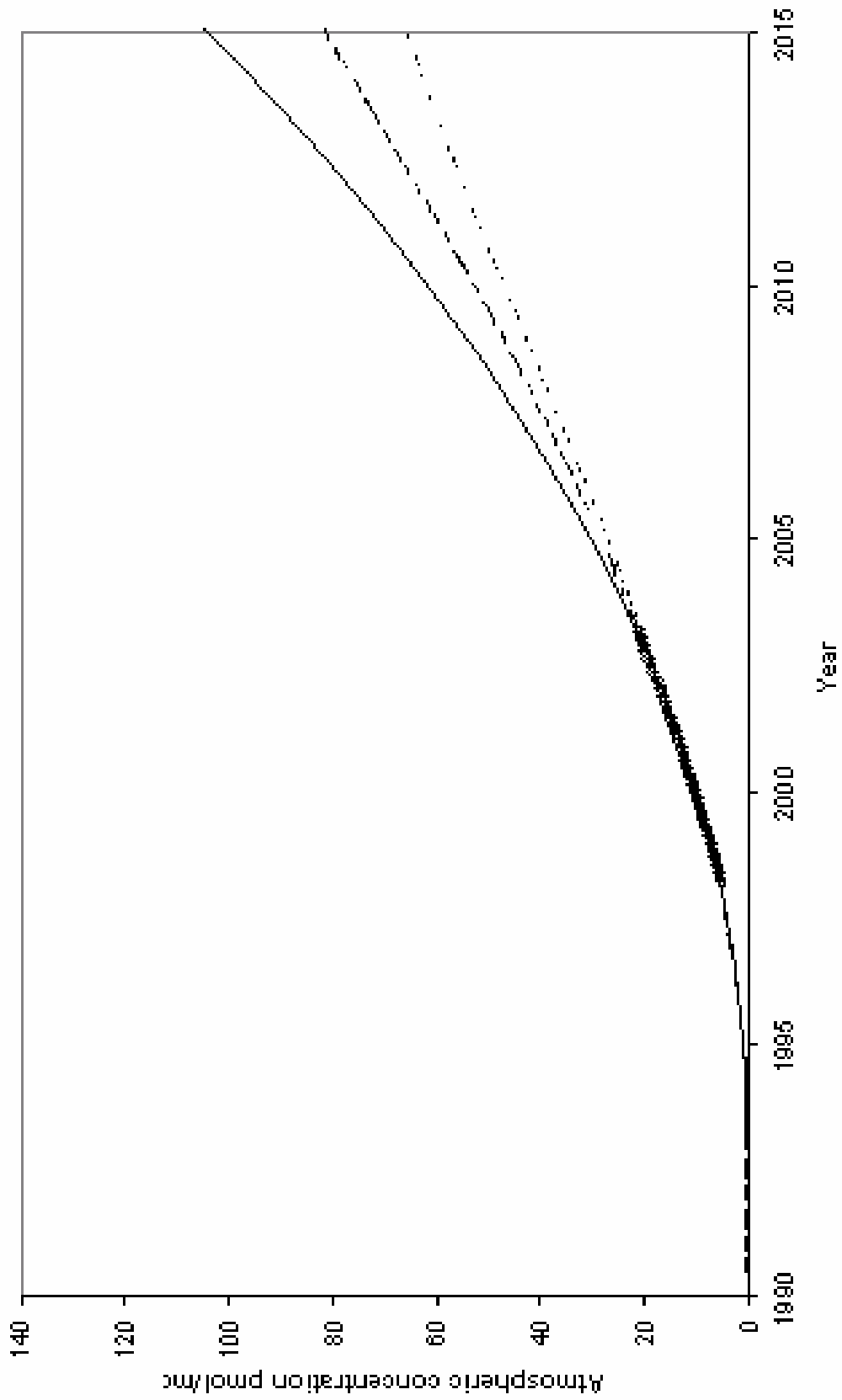
**Figure 8.** Atmospheric Concentrations of HFC-125. Open diamonds show the annual global average concentrations from Montzka, Fraser *et al.* (2003). Lines show the concentrations calculated from emissions for all three scenarios.

### HFC-134a

The atmospheric concentrations of HFC-134a are shown in Figures 9 and 10. For this compound, agreement between concentrations calculated from emissions in Table 3.2 and the measurements from AGAGE (2004) is striking. Counting statistics for the measurements are similar to those for HCFCs 141b and 142b (75 measurements per data point) and the average coefficient of variance is 2.5% thus there is no statistical difference between the measurements and the calculated concentrations.



**Figure 9.** Northern hemispherical concentrations of HFC-134a. Plus signs indicate monthly means of measurements at Mace Head, Ireland. Lines are calculated concentrations using all three scenarios.



**Figure 10.** Southern hemispherical concentrations of HFC-134a showing measurements at Cape Grim, Tasmania and calculated concentrations as in Figure 9.

### **HFC-143a**

There are no reported atmospheric measurements, so the expected concentrations shown in Figure 11 rely solely on the prognosis for emissions shown in Tables 3.2 and 3.7 to 3.9.

### **HFC-152a**

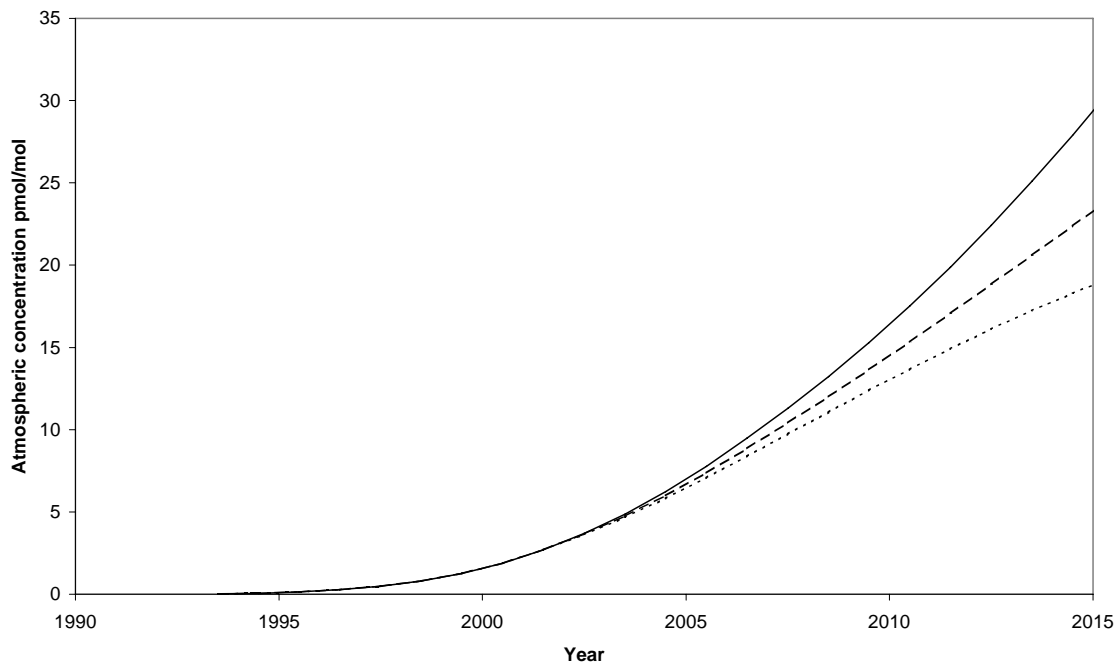
Concentrations of this substance are dominated by the prompt emissions from aerosols and open cell foams as reported in UNFCCC (2003). In the absence of better data, these emissions were fitted (together with those estimated expressly for refrigeration and closed cell foams) to the atmospheric concentrations measured. An exact fit, as shown in Figure 12, was obtained with a growth rate of prompt emissions of 1250 tonnes/year/year starting in 1990.

### **Minor HFCs - 227ea, 245fa and 365mfc**

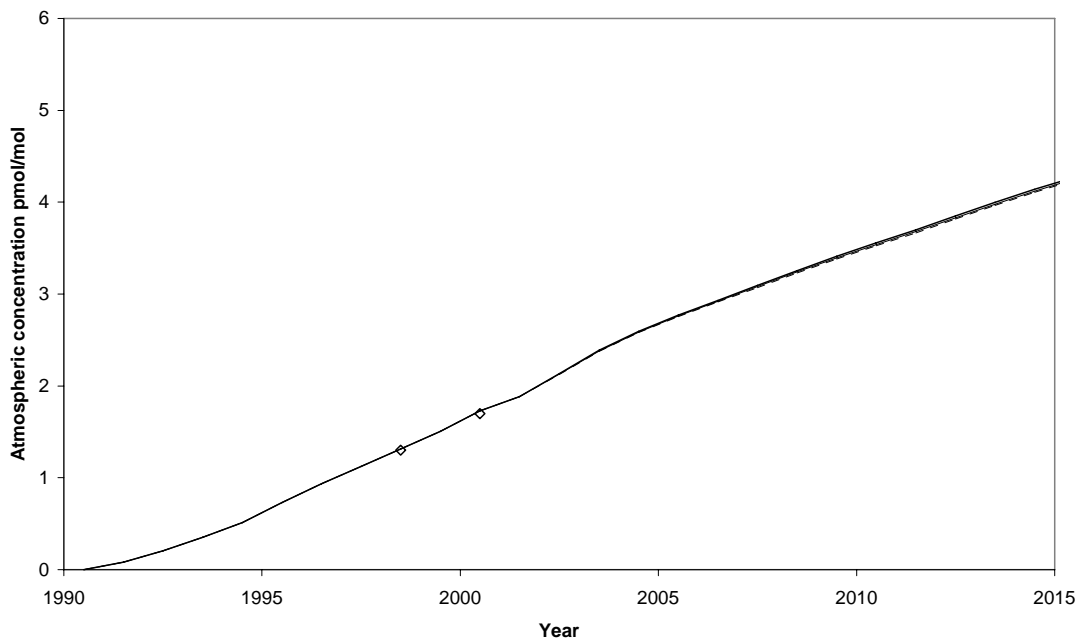
Calculated atmospheric concentrations for one scenario for emissions of these are shown in Figure 13; there are no measurements reported yet.

### **HFC-32**

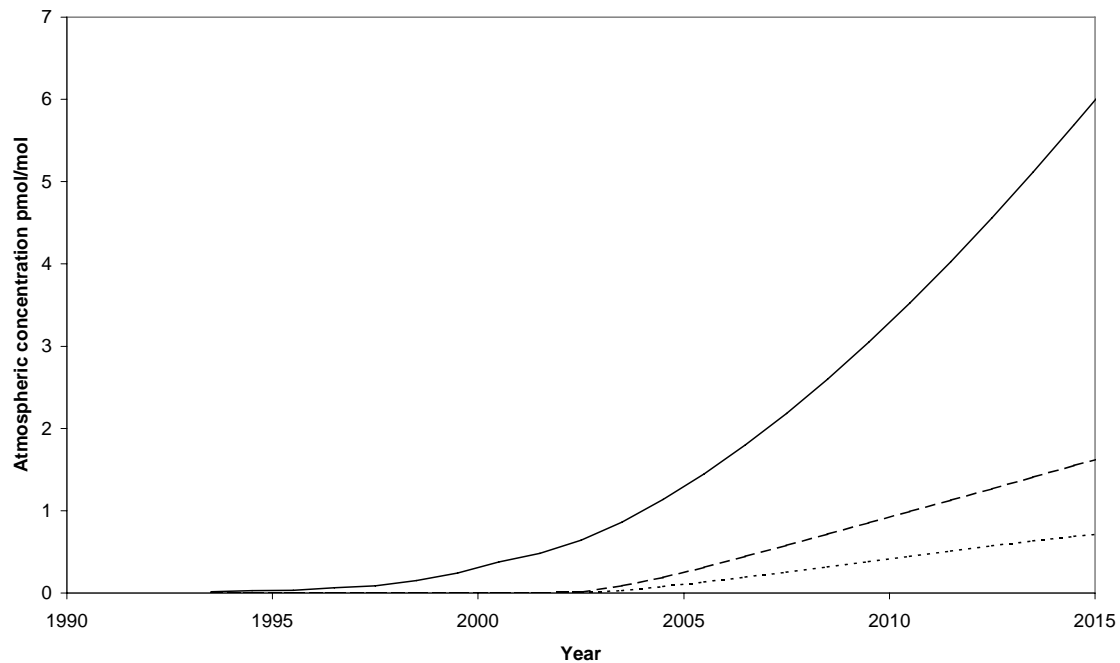
Again, no measurements of this have been reported and Figure 14 shows the calculated globally averaged concentrations, for all scenarios.



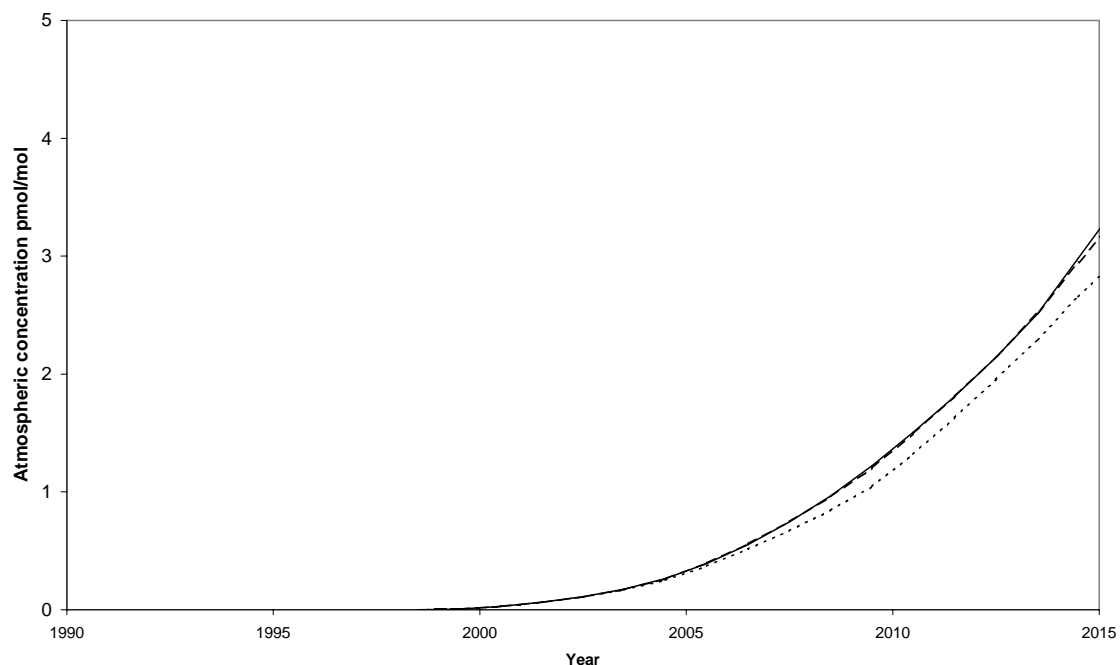
**Figure 11.** Calculated concentrations of HFC-143a for all scenarios. Solid line - Scenario1, dashed line - Scenario 2 and dotted line - Scenario 3.



**Figure 12.** Globally averaged atmospheric concentrations of HFC-152a. Open diamonds are the annual average concentrations from Montzka, Fraser *et al.* (2003). Lines are concentrations calculated from the emissions estimates shown in Tables 3.7 to 3.9 for all scenarios.



**Figure 13.** Calculated atmospheric concentrations of the minor HFCs: solid line - HFC-227ea, dashed line - HFC-245fa and dotted line - HFC-365mfc, based on the data in Tables 3.2 and 3.7. Only one scenario shown.



**Figure 14.** Calculated global average concentrations of HFC-32 for all scenarios based on emissions in Tables 3.2 and 3.7 to 3.9. Scenarios 1 - solid line, 2 - dashed line and 3 - dotted line.

## 4.2 Differences Between Calculations and Measurements

There are clear differences between calculated and observed concentrations for some, but not all of the fluorocarbons considered here and, while analysis of these differences for individual compounds is helpful, the difference in behaviour between the compounds may be instructive. The basic measurement and calculation methods are the same for all of the substances but an explanation is required for example for the failure of the calculated concentrations of CFC-12 in recent years to fit observations while those for HFC-134a fit almost exactly.

Possible factors contributing to the differences are: systematic errors in measurement that affect the observed atmospheric concentrations of some of the compounds, similar errors in some of the atmospheric lifetimes used to calculate concentrations, changes in the atmospheric lifetimes due to changes in atmospheric chemistry, systematic errors in activity used to calculate emissions, similar errors in the emission functions to convert activity into the time series of emissions and changes in emission functions with time (as containment practices change).

A number of these can be ruled out:

*Systematic errors in the measurements* would have to be duplicated across several laboratories. The intercomparisons that are carried out routinely have explained the differences that exist between the determinations as consequences of, for example, the use of different standards or the use of different statistical algorithms to separate results from polluted air from background samples. The possibility that the discrepancies between calculations and measurements that exist for example for HCFC-141b and HCFC-142b is the result of some systematic analytical error that affects all laboratories equally is vanishingly small.

*Systematic errors in atmospheric lifetimes* are equally unlikely. These are calculated using mathematical models of atmospheric chemistry and physics and the results are the subject of periodic review under the WMO Assessments of stratospheric ozone depletion, the latest being Montzka, Fraser *et al.* (2003). Atmospheric lifetimes have uncertainties in the region of 10% and any changes in the assessed values have been within this range. Furthermore, the atmospheric lifetime will affect the whole time series of the calculation and cannot account for points of inflection in the calculations (as observed for CFCs 11, 12 and 115 and HCFC-22).

*Changes in atmospheric lifetimes* can occur on a time scale of years to decades. However, these are cyclical with the largest effect arising from seasonal changes in the oxidising power of the atmosphere as the solar angle changes (affecting short lived compounds such as HCFC-123 and HFC-152a most). The 11-year solar cycle has a minor effect on compounds that decompose in the stratosphere but, again, this effect is cyclical and would not account for points of inflection.

The remaining potential sources of the differences merit further work beyond the scope of this report. The database of activities before 1990 is different from that developed here and is coincident with the points of inflection in the calculated concentration records. Similarly, the emission functions to convert this activity into the time series of emissions are different before 1990. One interesting speculation is that, as containment practices have changed (which has undoubtedly happened), these emission functions may have changed, so that

application of a single parameter to the whole time series of activity may not be appropriate. However, deconvoluting the influences of these factors will require significant new effort.

Nevertheless, even where there are significant differences between measurements and calculation, examination of the effect of future scenarios is worthwhile, to draw relative conclusions, which is the case for CFCs. For the HFCs, the calculated concentrations are a better match for observations and absolute conclusions may be drawn.

### **4.3 Future Atmospheric Concentrations**

#### **CFC-11 and CFC-12**

Despite the absolute difference between calculated and measured concentrations, the differences between the scenarios for future emissions have relatively little effect on atmospheric concentrations. This is mainly due to the fact that the concentrations are governed by the rate of removal of the current atmospheric burdens (5 million tonnes of CFC-11 and almost 10 million tonnes of CFC-12) so that the relatively small amounts predicted to be released in the future have a similarly small effect.

#### **CFC-115**

The future concentration of CFC-115 depends on forecast emissions that would require an additional 120000 tonnes to be produced between 2001 and 2015. Given that the annualised rate would be close to the historic maximum production (in the developed world) and that most of the future material would have to be produced by (or for) the developing world, ceasing in 2010, the forecast does not seem viable. However, the absolute concentrations, and their contribution to stratospheric ozone depletion and climate change, are small.

#### **HCFC-123 and HCFC-124**

Anticipated concentrations of HCFCs 123 and 124 for Scenario 1 are shown in Figure 6. In neither case does the concentration exceed  $1 \text{ pmol mol}^{-1}$  and so are inconsequential.

#### **HCFC-141b and HCFC-142b**

The difference between calculated and observed concentrations of these compounds is so large that prediction based on the calculated values is unsound. However, this is mitigated by the fact that, even based on the measured concentrations, forecast contributions to total HCFC concentrations in the atmosphere (in the region of  $50 \text{ pmol mol}^{-1}$ ) are much smaller than those arising from HCFC-22.

#### **HCFC-22**

Figure 7 shows the future concentrations anticipated from the scenarios; in all cases there is a significant increase from about  $150 \text{ pmol mol}^{-1}$  now, to within the range from 200 to  $300 \text{ pmol mol}^{-1}$  by 2015. Measured concentrations are relatively well matched by those calculated from historic emissions data and so the uncertainty of the forecast concentrations is mainly a consequence of scenario uncertainty.



## **HFC-125**

Atmospheric measurements of the concentration of HFC-125 are substantially higher than those calculated from historic emissions (Montzka, Fraser *et al.*, 2003). However, the absolute magnitude of the concentrations is very low (less than 2 pmol mol<sup>-1</sup>) and so the measurement errors may be substantial. There is no indication of a significant source of emissions (such as aerosol propellant) other than refrigeration and so the expected growth in atmospheric concentration for all scenarios shown in Figure 8 is likely to be representative.

## **HFC-134a**

The atmospheric concentrations of HFC-134a are shown in Figures 9 and 10. The atmospheric concentration of HFC-134a is likely to remain predominant among HFCs up to 2015, on all scenarios, with a minimum concentration of 60 pmol mol<sup>-1</sup> in the southern hemisphere and a maximum of 130 pmol mol<sup>-1</sup> in the northern in 2015.

## **HFC-143a**

By 2015, this is likely to be the second most abundant of the HFCs used as ODS substitutes, with concentrations in the range 17 to 30 pmol mol<sup>-1</sup>, as shown in Figure 11. There are no reported atmospheric measurements, so this expectation relies solely on the prognosis for emissions.

## **HFC-152a**

There is no difference between the atmospheric concentrations arising from the scenarios for HFC-152a, Figure 12. The scenarios apply to use in refrigeration but these concentrations are dominated by the prompt emissions from aerosols and open cell foams for which only one, rather arbitrary, scenario has been adopted.

## **HFCs - 227ea, 245fa and 365mfc**

Emissions of the minor HFCs - 227ea, 245fa and 365mfc - have not been ascribed scenarios. In the case of the first, this is because most of the projected growth in emissions is in firefighting, which is out of the scope of this study, and for the second two, the absolute level of emissions is very small, so that concentrations barely exceed 1 pmol mol<sup>-1</sup> by 2015. These are shown in Figure 13.

## **HFC-32**

Unlike HFC-152a, the lack of difference between atmospheric concentrations arising from scenarios 1 and 2 for HFC-32 is a consequence of the similarity between the scenarios (see Figure 14). Only with the more marked reduction in emissions of scenario 3 is there a lessening in the growth rate of atmospheric concentration.

## 5. Effect of Future Atmospheric Concentrations of HFCs on Climate Change

The effect on climate that can potentially arise from the materials considered here arises from their ability to absorb infrared radiation and their propensity to accumulate in the atmosphere. The HFCs are greenhouse gases that are included in the Kyoto Protocol; CFCs and HCFCs are also greenhouse gases but, because they are already controlled under the Montreal Protocol, as a consequence of ozone depletion, they are not regulated under Kyoto. The potential climate change impact of an emission can be given a numerical ranking relative to carbon dioxide by multiplying the mass emission by the relative "Global Warming Potential" (GWP) of the compound. The latter is a conversion factor and the units of the result are mass of CO<sub>2</sub> equivalent. Such calculations have been performed for the emissions from use in refrigeration in Section 1 of this report.

However, the product of mass and GWP conversion factor only gives the total impact over the next 100 years (in terms of the total impact over the same time of an equivalent quantity of carbon dioxide). The actual impact and its timing would be of more value. One measure of this impact is the expected "radiative forcing" of climate, which is an absolute quantity with units of watts per square metre which can be directly used in climate models to predict temperature changes. While detailed modelling is required to predict exact temperatures, each 1°C requires approximately 2 Wm<sup>-2</sup>. For low gas concentrations (such as for the HFCs) and long pathlengths (as in the atmosphere), the absorption is nearly proportional to concentrations. So that, in cases where the time series of atmospheric concentrations of the greenhouse gases is available, a close approximation to the development of their radiative forcing in time can be calculated. At any point in time, it is the product of their concentration and a radiative forcing constant, the units of which are watts per square metre per concentration unit (generally parts per billion - ppb - or nanomol mol<sup>-1</sup>). The radiative forcing constant is also known as the Absolute Global Warming Potential but this nomenclature is not used here to avoid confusion.

The development of radiative forcing up to 2015 for all HFC emissions (plus HFC-23, which is not a subject of this report but makes a significant contribution) is shown in Figure 15. This uses the forcing constants from Houghton *et al.* (2001) shown in Table 5.1.

**Table 5.1.** Radiative Forcing Constants for the Common HFCs

Substance	Radiative forcing constant W m <sup>-2</sup> ppb <sup>-1</sup>	Substance	Radiative forcing constant W m <sup>-2</sup> ppb <sup>-1</sup>
HFC-125	0.23	HFC-245fa	0.28
HFC-134a	0.15	HFC-32	0.09
HFC-143a	0.13	HFC-365mfc	0.21
HFC-152a	0.09	HFC-23	0.16
HFC-227ea	0.3		

While the rate of increase in the impact from HFCs appears large from Figure 15, the absolute value is less than 40 milliwatts per square metre in 2015 and it is necessary to place this into the context of the impact of all greenhouse gases.

The basis of Scenario 1 in this report, in which existing environmental legislation is applied but there are few new constraints on greenhouse gas emissions, is similar to the background for the scenario designated B2 in the Special Report on Emissions Scenarios (SRES) compiled for IPCC (Nakicenovic *et al.*, 2000). This scenario provides forecasts for all greenhouse gases up to the year 2100 for a future in which environmental legislation is imperfectly applied, and there are no new draconian constraints on emissions but also steady, rather than explosive, economic growth.

Figure 16 shows the radiative forcing arising from the B2 scenario for carbon dioxide, methane, nitrous oxide, perfluorocarbons, sulphur hexafluoride, and the ozone depleting substances up to 2015. The HFC emissions are from Scenario 1 in this report.

While the impact from HFCs up to 2015 is discernible, it is not a significant part of the global impact which is dominated by carbon dioxide.

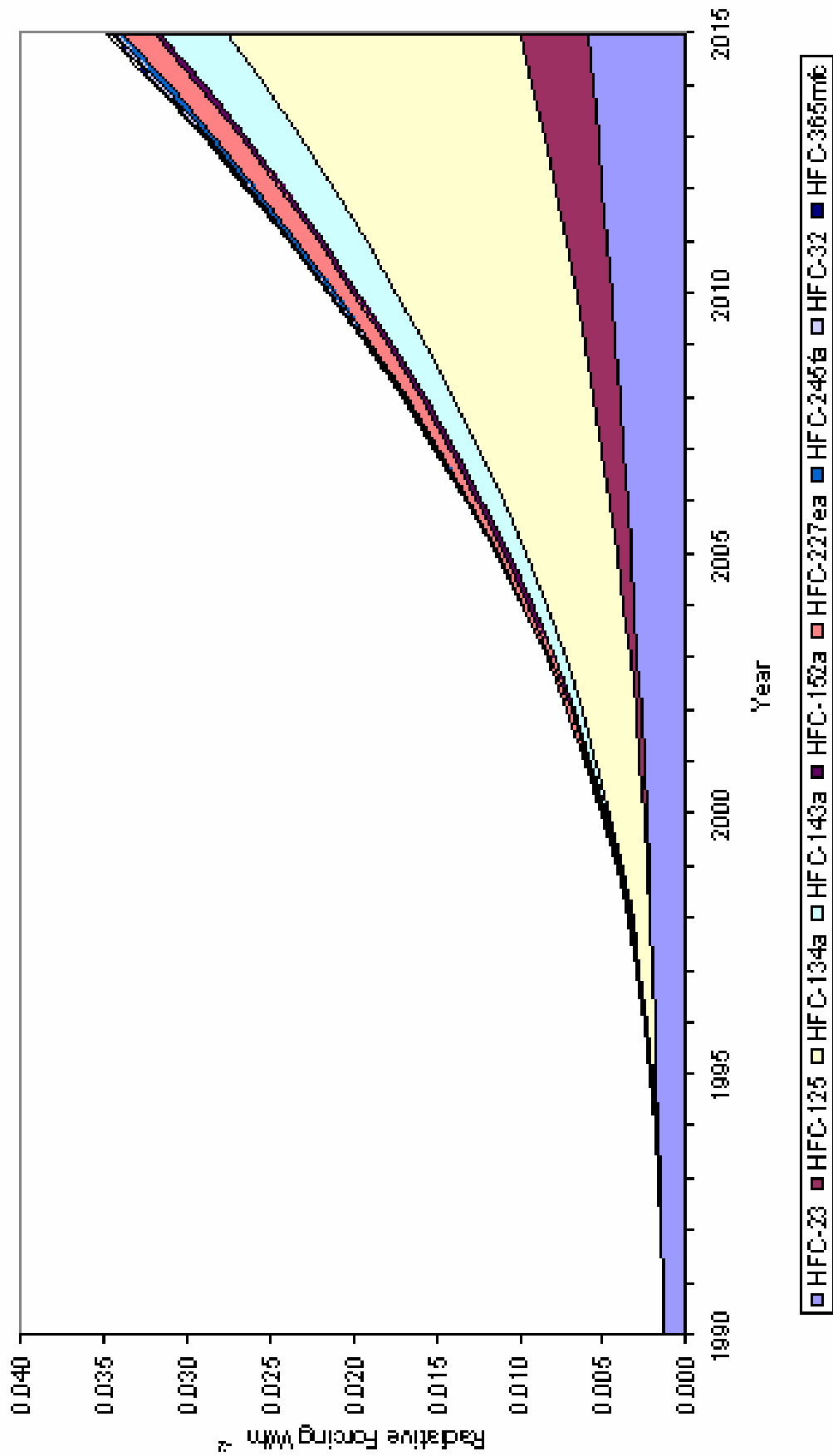
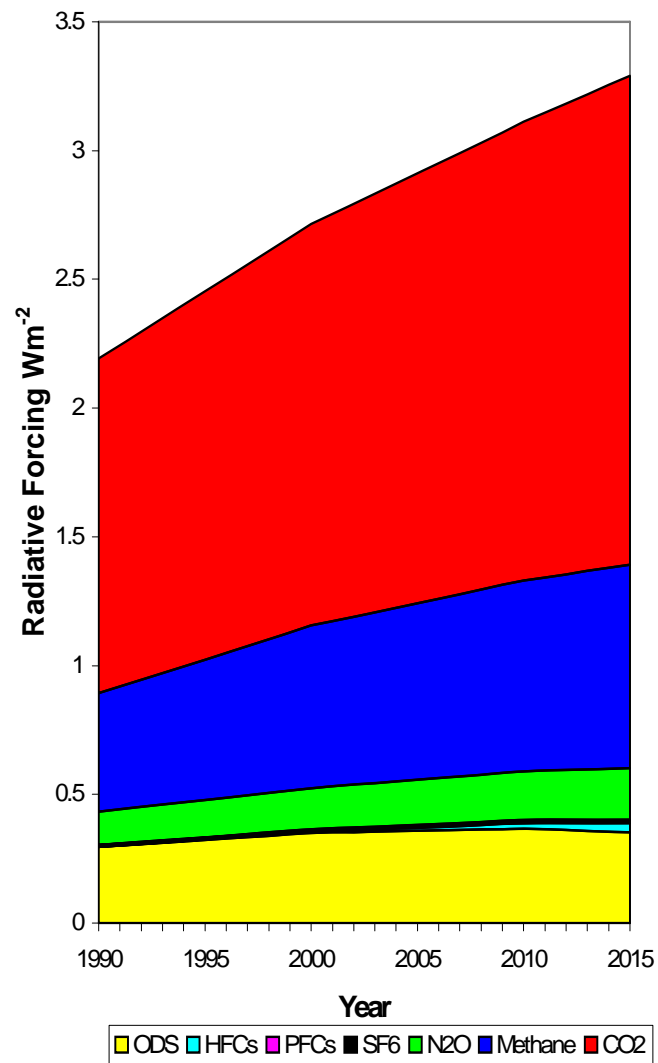


Figure 15. Radiative Forcing from HFCs over the years to 2015



**Figure 16.** Future Impact of Emissions of all Greenhouse Gases. Summation of contributions calculated to commence in year 1760 (to give a value for the total anthropogenic effect).

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